

I. Basis of the opinion**1. With regard to the elements of the international application:***

- ☒ the international application as originally filed
- ☒ the description:
pages 1-30 _____, as originally filed
pages NONE _____, filed with the demand
pages NONE _____, filed with the letter of _____
- ☒ the claims:
pages 31-34 _____, as originally filed
pages NONE _____, as amended (together with any statement) under Article 19
pages NONE _____, filed with the demand
pages NONE _____, filed with the letter of _____
- ☐ the drawings:
pages NONE _____, as originally filed
pages NONE _____, filed with the demand
pages NONE _____, filed with the letter of _____
- ☐ the sequence listing part of the description:
pages NONE _____, as originally filed
pages NONE _____, filed with the demand
pages NONE _____, filed with the letter of _____

2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item. These elements were available or furnished to this Authority in the following language _____ which is:

- ☐ the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of the translation furnished for the purposes of international preliminary examination (under Rules 55.2 and/or 55.3).

3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the written opinion was drawn on the basis of the sequence listing:

- ☐ contained in the international application in printed form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. ☐ The amendments have resulted in the cancellation of:

- ☐ the description, pages NONE _____
- ☐ the claims, Nos. NONE _____
- ☐ the drawings, sheets/fig NONE _____

5. ☐ This opinion has been drawn as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).

* Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this opinion as "originally filed."

WRITTEN OPINION

International application No.
PCT/ 6312

V. Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. STATEMENT

Novelty (N)	Claims <u>12 and 13</u>	YES
	Claims <u>1-11 and 14-23</u>	NO
Inventive Step (IS)	Claims <u>12 and 13</u>	YES
	Claims <u>1-11 and 14-23</u>	NO
Industrial Applicability (IA)	Claims <u>1-23</u>	YES
	Claims <u>NONE</u>	NO

2. CITATIONS AND EXPLANATIONS

Claims 1-11, and 14-23 lack novelty under PCT Article 33(2) as being anticipated by Chu et al (5,516,812 A) (D1) and lacks novelty under PCT Article 33(3) as being obvious over Chu et al (5,516,812 A) (D1).

Chu et al teaches UV-moisture curable silicone conformal coatings. Said coating comprises a silicone fluid of a monovalent ethylenically unsaturated functional group end-capped silicone, at least one (meth) acryl-functionalized silicone, and a photoinitiator. Said monovalent ethylenically unsaturated functional group end-capped silicone is a reaction product obtained by a silanol-terminated silicone and a silane crosslinker. Said crosslinker has directly adjoined to a silicone atom a monovalent ethylenically unsaturated functional group and at least 2 hydrolyzable groups, which reads on formula I in claims 1 and 2, wherein R1 is a hydrolyzable group. Said silanol-terminated silicone is a linear polyorganosilane, which reads on formula II in claim 1 and formula IV in claim 2. Said crosslinkers is used in amounts from 1 to 50% by weight based on the silanol-terminated silicone. When the viscosity of the composition needs to be modified, Chu et al teaches adding non-reactive silicones-column 8, lines 45-52. Said (meth) acryl-functionalized silicone can be compounds, such as (meth) acryloxypropyldimethoxysilyl-terminated silicones.

Chu et al teaches that the composition can be cured using actinic radiation and moisture, in cases where the coating is shaded from the actinic radiation exposure. Chu et al teaches that elevated temperatures can be employed to cure the composition to speed up the reaction rate. When curing with actinic radiation, photoinitiators are used in the composition. Photoinitiators, such as those list in column 9, lines 8-22 can be used, wherein diethoxyacetophenone is listed. Photoinitiators can be added in amounts from 0.1% to 15% by weight based on the curable components in the composition. Moisture cure initiators, such as orthotitanates can be added to the composition. Chu et al does not teach effective amounts, however it is customary to added only small amounts, such as 0.1 to 5%, of initiators, as suggested by the recommended amounts of photoinitiators. The (meth) acryl-functionalized silicone can be added in amounts from 80 to about 60 parts by weight. The ethylenically unsaturated functional group end-capped silicone can be added to the composition in amounts from 20 to about 35 parts by weight of the composition.

D1 teaches the claimed invention, therefore the instant invention is deemed to lack novelty in view of the prior art.

Claims 1-23 meet industrial applicability as defined by PCT Article 33(4). The compositions and methods taught by the instant invention can be useful as a conformal coating.

Claims 12-13 the criteria set out in PCT Article 33(2)-(4), because the prior art does not teach or fairly suggest using vinyl terminated silicone resins or organosiloxane resin as reactive diluents in composition of claim 1 nor is the mol% of claim 13 taught by the reference or the prior art.

----- NEW CITATIONS -----

WRITTEN OPINION

International application No.
PCT/JP00/16312

Supplemental Box

(To be used when the space in any of the preceding boxes is not sufficient)

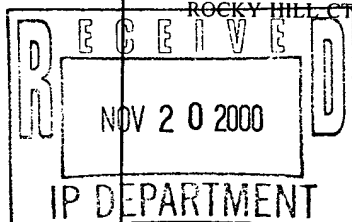
TIME LIMIT:

The time limit set for response to a Written Opinion may not be extended. 37 CFR 1.484(d). Any response received after the expiration of the time limit set in the Written Opinion will not be considered in preparing the International Preliminary Examination Report.

PATENT COOPERATION TREATY

From the INTERNATIONAL SEARCHING AUTHORITY

To: STEVEN C. BAUMAN
LOCTITE CORPORATION
1001 TROUT BROOK CROSSING
ROCKY HILL, CT 06067



PCT

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL SEARCH REPORT OR THE DECLARATION

(PCT Rule 44.1)

Applicant's or agent's file reference LC-366-PCT	Date of Mailing (day/month/year) 14 NOV 2000
International application No. PCT/US00/16312	International filing date (day/month/year) 07 JULY 2000
Applicant LOCTITE CORPORATION	

1. ☒ The applicant is hereby notified that the international search report has been established and is transmitted herewith.
Filing of amendments and statement under Article 19:
 The applicant is entitled, if he so wishes, to amend the claims of the international application (see Rule 46):

When? The time limit for filing such amendments is normally 2 months from the date of transmittal of the international search report; however, for more details, see the notes on the accompanying sheet.
Where? Directly to the International Bureau of WIPO
 34, chemin des Colombettes
 1211 Geneva 20, Switzerland
 Facsimile No.: (41-22) 740.14.35
For more detailed instructions, see the notes on the accompanying sheet.

2. ☐ The applicant is hereby notified that no international search report will be established and that the declaration under Article 17(2)(a) to that effect is transmitted herewith.

3. ☐ **With regard to the protest against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:**

☐ the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.
☐ no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.

4. **Further action(s):** The applicant is reminded of the following:

Shortly after **18 months** from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in rules 90 *bis* 1 and 90 *bis* 3, respectively, before the completion of the technical preparations for international publication.
 Within **19 months** from the priority date, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later).
 Within **20 months** from the priority date, the applicant must perform the prescribed acts for entry into the national phase before all designated Offices which have not been elected in the demand or in a later election within 19 months from the priority date or could not be elected because they are not bound by Chapter II.

Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703) 305-3230	Authorized officer SANZA MCCLENDON Telephone No. (703) 308-0651 <div style="text-align: right;"> DEBORAH THOMAS <i>Det</i> PARALEGAL SPECIALIST </div>
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PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference LC-366-PCT	FOR FURTHER ACTION see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. PCT/US00/16312	International filing date (day/month/year) 07 JULY 2000	(Earliest) Priority Date (day/month/year) 19 JULY 1999
Applicant LOCTITE CORPORATION		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 3 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing:

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

2. ☐ **Certain claims were found unsearchable** (See Box I).

3. ☐ **Unity of invention is lacking** (See Box II).

4. With regard to the **title**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No. _____

☐ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

☐ None of the figures.

REC'D 19 FEB 2002

WIPO PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

10

Applicant's or agent's file reference LC-366-PCT	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/US00/16312	International filing date (day/month/year) 07 July 2000 (07.07.2000)	Priority date (day/month/year) 19 July 1999 (19.07.1999)
International Patent Classification (IPC) or national classification and IPC IPC(7): C08F 2/48; C08L 67/00; 75/06; C08G 63/00 and US Cl.: 522/60; 24, 66, 71, 81, 104, 105.; 528/280, 281, 282; 525/410, 411, 437, 445		
Applicant LOCTITE CORPORATION		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
 2. This REPORT consists of a total of 3 sheets, including this cover sheet.
- ☒ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 2 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of report with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand 20 February 2001 (20.02.2001)	Date of completion of this report 13 January 2002 (13.01.2002)
Name and mailing address of the IPEA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703)305-3230	Authorized officer James Seidleck Telephone No. (703) 308-0657

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International Application No. _____
PCT/US00/____2

I. Basis of the report

1. With regard to the elements of the international application:*

☐ the international application as originally filed.

☒ the description:

pages 1-3, 6-9, 11-30 as originally filed

pages NONE, filed with the demand

pages 4, 5 and 10, filed with the letter of 01 October 2001 (01.10.2001)

☒ the claims:

pages NONE, as originally filed

pages NONE, as amended (together with any statement) under Article 19

pages NONE, filed with the demand

pages 31-34, filed with the letter of 01 October 2001 (01.10.2001)

☐ the drawings:

pages NONE, as originally filed

pages NONE, filed with the demand

pages NONE, filed with the letter of _____.

☐ the sequence listing part of the description:

pages NONE, as originally filed

pages NONE, filed with the demand

pages NONE, filed with the letter of _____.

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language _____ which is:

☐ the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).

☐ the language of publication of the international application (under Rule 48.3(b)).

☐ the language of the translation furnished for the purposes of international preliminary examination (under Rules 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

☐ contained in the international application in printed form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. ☐ The amendments have resulted in the cancellation of:

☐ the description, pages NONE

☐ the claims, Nos. NONE

☐ the drawings, sheets/fig NONE

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).**

* Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17).

** Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International Application No.
PCT/US

V. Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. STATEMENT

Novelty (N)	Claims <u>1-23</u>	YES
	Claims <u>NONE</u>	NO
Inventive Step (IS)	Claims <u>1-23</u>	YES
	Claims <u>NONE</u>	NO
Industrial Applicability (IA)	Claims <u>1-23</u>	YES
	Claims <u>NONE</u>	NO

2. CITATIONS AND EXPLANATIONS

Claims 1-23 meet the criteria set out in PCT Article 33(2)-(4), because the prior art does not teach or fairly suggest a resin reinforced silicone composition curable by exposure to radiation or exposure to moisture or a combination, thereof, comprising (a) a polyorganosiloxanes with reactive groups selected from moisture curable groups, photocurable groups, or combinations thereof; (b) a silicone resin selected from the resins in claim 1 or 2; and (c) a cure catalyst selected from the groups consisting of photoinitiators, moisture cure catalyst of combinations thereof. Nor does the prior art teach a method of curing the above compositions to form an elastomer comprising the steps of claims 21-23. The prior art document (D1) fails to teach the silicone resins as defined and used in applicant's invention which, are three-dimensional structures verses the linear silicone fluids taught in D1. Additionally, D1 fails to teach using said silicone fluids to reinforce polyorganosiloxanes. The compositions and methods taught by the instant invention can be useful as a conformal coating.

----- NEW CITATIONS -----

REGARDING THE INTERNATIONAL APPLICATION OF
DOCKET OR REFERENCE NUMBER: LC-366/PCT
LOCTITE CORPORATION, and CHU, H. H.

ENTITLED

Resin-Reinforced UV, Moisture and UV/Moisture Dual Curable Silicone Compositions

Certification under 37 CFR 1.10 (if applicable)

"Express Mail" mailing number

Date of Deposit

I hereby certify that this application is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Commissioner of Patents and Trademarks, Box PCT, Washington, D.C. 20231.

(Typed or printed name of person
mailing application)(Signature of person mailing
application)

To the United States Receiving Office (RO/US):

Accompanying this transmittal letter is the above-identified International application, including a completed Request form (PCT/RO/101). Please process the application according to the provisions of the Patent Cooperation Treaty.

The following requests are made of the RO/US:

1. ☒ PREPARATION AND TRANSMITTAL OF CERTIFIED COPY OF PRIORITY DOCUMENTS - Please prepare and transmit to the International Bureau a certified copy of the United States origin priority documents identified in Box VI of the Request form (37 CFR 1.451).

To cover the cost of copy preparation and certification (37 CFR 1.19(a)(3) and (b)(1),

- ☐ a (check) (money order) in the amount of \$ _____ is attached to this transmittal letter.
☒ the RO/US is hereby authorized to charge the following deposit account no.: 12-2135.

The appropriate Search fee for the above-named Authority is indicated on the Fee Calculation Sheet (PCT/RO/101 Annex).

2. ☒ SUPPLEMENTAL SEARCH FEES (ONLY WHEN ISA/US CONDUCTS THE INTERNATIONAL SEARCH) - Please charge any Supplemental Search fees that may be required by the United States International Searching Authority (ISA/US) to deposit account no.: 12-2135.

I understand that this authorization is subject to my oral confirmation thereof in each instance and that it in no way limits my right to submit a protest against payment of the Supplemental Search fees, but is merely an administrative aid to assure that the ISA/US may timely complete the Search Report.

NOTE: SUPPLEMENTAL SEARCH FEES FOR ISA/EP ARE PAYABLE DIRECTLY TO THE EUROPEAN PATENT OFFICE

3. ☒ DISCLOSURE INFORMATION - In order to assist in screening the accompanying International application for purposes of determining whether a license for foreign transmittal should and could be granted and for other purposes, the following information is supplied:
- A. ☐ There is no prior filed application relating to this invention.
- B. ☒ There is a prior application*, serial number 60/144,257 filed on July 19, 1999 which contains subject matter that is
1. ☐ substantially identical to that of the accompanying International application.
 2. ☒ less than that of the accompanying International application. The additional subject matter of the International application appears on page(s) and line(s) throughout the application.
 3. ☐ more than that of the accompanying International application.
- C. ☐ Disclosure information cannot be covered by the language of Points 3A or 3B above due to the involvement of several prior applications or for other reasons. A separate sheet on which the disclosure information is explained is attached to this transmittal letter.
4. ☒ REQUEST FOR FOREIGN TRANSMITTAL LICENSE - According to the provisions of 35 U.S.C. 184 and 37 CFR 5.11, a license to transmit the accompanying International application to foreign agencies or International authorities is hereby requested.

*Priority is not claimed, unless all necessary information is listed in Box VI of the Request Form (PCT/RO/101).

SIGNER IS THE:

- ☐ APPLICANT
☐ COMMON REPRESENTATIVE
☒ (ATTORNEY) (AGENT)
REG. NO. 33,832

NAME OF SIGNER (typed)

Steven C. Bauman

SIGNATURE

PCT COOPERATION TREATY

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Commissioner
 US Department of Commerce
 United States Patent and Trademark
 Office, PCT
 2011 South Clark Place Room
 CP2/5C24
 Arlington, VA 22202
 ETATS-UNIS D'AMERIQUE
 in its capacity as elected Office

Date of mailing (day/month/year) 25 April 2001 (25.04.01)	
International application No. PCT/US00/16312	Applicant's or agent's file reference LC-366-PCT
International filing date (day/month/year) 07 July 2000 (07.07.00)	Priority date (day/month/year) 19 July 1999 (19.07.99)
Applicant CHU, Hsien-Kun	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:
 20 February 2001 (20.02.01)

☐ in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was
☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35	Authorized officer Claudio Borton Telephone No.: (41-22) 338.83.38
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PCT REQUEST

1/4

Original (for SUBMISSION) - printed on 06.07.2000 03:47:04 PM

LC-366-PCT

0	For receiving Office use only	
0-1	International Application No.	
0-2	International Filing Date	
0-3	Name of receiving Office and "PCT International Application"	
0-4	Form - PCT/RO/101 PCT Request	
0-4-1	Prepared using	PCT-EASY Version 2.90 (updated 08.03.2000)
0-5	Petition The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty	
0-6	Receiving Office (specified by the applicant)	United States Patent and Trademark Office (USPTO) (RO/US)
0-7	Applicant's or agent's file reference	LC-366-PCT
I	Title of invention	RESIN-REINFORCED CURABLE SILICONE COMPOSITIONS
II	Applicant	
II-1	This person is:	applicant only
II-2	Applicant for	all designated States except US
II-4	Name	LOCTITE CORPORATION
II-5	Address:	1001 Trout Brook Crossing Rocky Hill, CT 06067 United States of America
II-6	State of nationality	US
II-7	State of residence	US
II-8	Telephone No.	(860) 571-5100
II-9	Facsimile No.	(860) 571-5465
II-10	e-mail	steve.bauman@loctite.com
III-1	Applicant and/or inventor	
III-1-1	This person is:	applicant and inventor
III-1-2	Applicant for	US only
III-1-4	Name (LAST, First)	CHU, Hsien-kun
III-1-5	Address:	6 Harvest Hill Wethersfield, CT 06109 United States of America
III-1-6	State of nationality	US
III-1-7	State of residence	US

PCT REQUEST

2/4

Original (for SUBMISSION) - printed on 06.07.2000 03:47:04 PM

LC-366-PCT

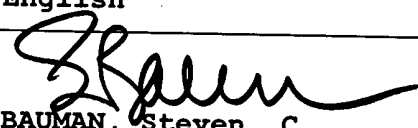
IV-1	Agent or common representative; or address for correspondence The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as:	agent BAUMAN, Steven, C. LOCTITE CORPORATION 1001 Trout Brook Crossing Rocky Hill, CT 06067 United States of America (860) 571-5001 (860) 571-5028 steve.bauman@loctite.com
IV-1-1	Name (LAST, First)	
IV-1-2	Address:	
IV-1-3	Telephone No.	
IV-1-4	Facsimile No.	
IV-1-5	e-mail	
V	Designation of States	
V-1	Regional Patent (other kinds of protection or treatment, if any, are specified between parentheses after the designation(s) concerned)	AP: GH GM KE LS MW SD SL SZ TZ UG ZW and any other State which is a Contracting State of the Harare Protocol and of the PCT EA: AM AZ BY KG KZ MD RU TJ TM and any other State which is a Contracting State of the Eurasian Patent Convention and of the PCT EP: AT BE CH&LI CY DE DK ES FI FR GB GR IE IT LU MC NL PT SE and any other State which is a Contracting State of the European Patent Convention and of the PCT OA: BF BJ CF CG CI CM GA GN GW ML MR NE SN TD TG and any other State which is a member State of OAPI and a Contracting State of the PCT
V-2	National Patent (other kinds of protection or treatment, if any, are specified between parentheses after the designation(s) concerned)	AE AG AL AM AT AU AZ BA BB BG BR BY CA CH&LI CN CR CU CZ DE DK DM DZ EE ES FI GB GD GE GH GM HR HU ID IL IN IS JP KE KG KP KR KZ LC LK LR LS LT LU LV MA MD MG MK MN MW MX NO NZ PL PT RO RU SD SE SG SI SK SL TJ TM TR TT TZ UA UG US UZ VN YU ZA ZW
V-5	Precautionary Designation Statement In addition to the designations made under items V-1, V-2 and V-3, the applicant also makes under Rule 4.9(b) all designations which would be permitted under the PCT except any designation(s) of the State(s) indicated under item V-6 below. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit.	

PCT REQUEST

3/4

Original (for SUBMISSION) - printed on 06.07.2000 03:47:04 PM

LC-366-PCT

V-6	Exclusion(s) from precautionary designations	NONE	
VI-1	Priority claim of earlier national application		
VI-1-1	Filing date	19 July 1999 (19.07.1999)	
VI-1-2	Number	60/144,257	
VI-1-3	Country	US	
VI-2	Priority document request The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) identified above as item(s):	VI-1	
VII-1	International Searching Authority Chosen	United States Patent and Trademark Office (USPTO) (ISA/US)	
VII-2	Request to use results of earlier search; reference to that search		
VII-2-1	Date		
VII-2-2	Number		
VII-2-3	Country (or regional Office)	US	
VIII	Check list	number of sheets	electronic file(s) attached
VIII-1	Request	4	-
VIII-2	Description	30	-
VIII-3	Claims	4	-
VIII-4	Abstract	1	lc-366_pct.txt
VIII-5	Drawings	0	-
VIII-7	TOTAL	39	
	Accompanying items	paper document(s) attached	electronic file(s) attached
VIII-8	Fee calculation sheet	✓	-
VIII-16	PCT-EASY diskette	-	diskette
VIII-17	Other (specified):	Return Receipt Postcard	-
VIII-18	Figure of the drawings which should accompany the abstract		
VIII-19	Language of filing of the international application	English	
IX-1	Signature of applicant or agent		
IX-1-1	Name (LAST, First)	BAUMAN, Steven, C.	

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10-1	Date of actual receipt of the purported international application	
10-2	Drawings:	
10-2-1	Received	
10-2-2	Not received	
10-3	Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application	
10-4	Date of timely receipt of the required corrections under PCT Article 11(2)	
10-5	International Searching Authority	ISA/US

PCT REQUEST

4/4

LC-366-PCT

Original (for SUBMISSION) - printed on 06.07.2000 03:47:04 PM

10-6	Transmittal of search copy delayed until search fee is paid	
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11-1	Date of receipt of the record copy by the International Bureau	
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PCT (ANNEX - FEE CALCULATION SHEET)

LC-366-PCT

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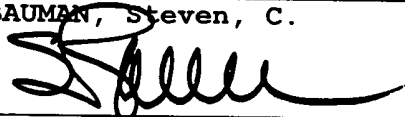
(This sheet is not part of and does not count as a sheet of the international application)

0	For receiving Office use only		
0-1	International Application No.		
0-2	Date stamp of the receiving Office		
0-4	Form - PCT/RO/101 (Annex)		
0-4-1	PCT Fee Calculation Sheet Prepared using	PCT-EASY Version 2.90 (updated 08.03.2000)	
0-9	Applicant's or agent's file reference	LC-366-PCT	
2	Applicant	LOCTITE CORPORATION, et al.	
12	Calculation of prescribed fees	fee amount/multiplier	total amounts (USD)
12-1	Transmittal fee T	\Rightarrow	240
12-2	Search fee S	\Rightarrow	700
12-3	International fee Basic fee (first 30 sheets) b1	427	
12-4	Remaining sheets	9	
12-5	Additional amount (X)	10	
12-6	Total additional amount b2	90	
12-7	b1 + b2 = B	517	
12-8	Designation fees Number of designations contained in international application	85	
12-9	Number of designation fees payable (maximum 8)	8	
12-10	Amount of designation fee (X)	92	
12-11	Total designation fees D	736	
12-12	PCT-EASY fee reduction R	-132	
12-13	Total International fee (B+D-R) I	\Rightarrow	1,121
12-14	Fee for priority document Number of priority documents requested	1	
12-15	Fee per document (X)	15	
12-16	Total priority document fee P	\Rightarrow	15
12-17	TOTAL FEES PAYABLE (T+S+I+P)	\Rightarrow	2,076
12-19	Mode of payment	authorization to charge deposit account	
12-20	Deposit account instructions The receiving Office:	United States Patent and Trademark Office (USPTO) (RO/US)	
12-20-1	is hereby authorized to charge the total fees indicated above to my deposit account	<input checked="" type="checkbox"/>	
12-20-2	is hereby authorized to charge any deficiency or credit any over-payment in the total fees indicated above to my deposit account	<input checked="" type="checkbox"/>	

PCT (ANNEX - FEE CALCULATION SHEET)

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12-20-3	is hereby authorized to charge the fee for preparation and transmittal of the priority document to the International Bureau of WIPO to my deposit account	✓
12-21	Deposit account No.	12-2135
12-22	Date	06 July 2000 (06.07.2000)
12-23	Name and signature	BAUMAN, Steven, C. 

VALIDATION LOG AND REMARKS

13-2-6	Validation messages Contents	Yellow! The power of attorney or a copy of the general power of attorney will need to be furnished unless all applicants sign the request form.
		Green? The international application contains no drawings. Please verify.
13-2-7	Validation messages Fees	Green? Please confirm that fee schedule utilized is the latest available
13-2-8	Validation messages Payment	Green? Please ensure that you have a valid deposit account with the receiving Office selected.

PATENT COOPERATION TREATY

RJP
2/12/02

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:
STEVEN C. BAUMAN
LOCTITE CORPORATION
1001 TROUT BROOK CROSSING
ROCKY HILL, CT 06067

FEB 20 2002

PCT

NOTIFICATION OF TRANSMITTAL OF INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Rule 71.1)

Date of Mailing
(day/month/year)

14 FEB 2002

Applicant's or agent's file reference

LC-366-PCT

IMPORTANT NOTIFICATION

International application No.

International filing date (day/month/year)

Priority date (day/month/year)

PCT/US00/16312

07 July 2000 (07.07.2000)

19 July 1999 (19.07.1999)

Applicant

LOCTITE CORPORATION

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.
4. **REMINDER**

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices)(Article 39(1))(see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/US

Commissioner of Patents and Trademarks
Box PCT
Washington, D.C. 20231

Facsimile No. (703)305-3230

Form PCT/IPEA/416 (July 1992)

Authorized officer

Sanza McClendon

Telephone No. (703) 308-0657

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference LC-366-PCT	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/US00/16312	International filing date (day/month/year) 07 July 2000 (07.07.2000)	Priority date (day/month/year) 19 July 1999 (19.07.1999)
International Patent Classification (IPC) or national classification and IPC IPC(7): C08F 2/48; C08L 67/00; 75/06; C08G 63/00 and US Cl.: 522/60; 24, 66, 71, 81, 104, 105.; 528/280, 281, 282; 525/410, 411, 437, 445		
Applicant LOCTITE CORPORATION		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
 2. This REPORT consists of a total of 3 sheets, including this cover sheet.
- ☒ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 7 sheets.

3. This report contains indications relating to the following items:
 - I ☒ Basis of the report
 - II ☐ Priority
 - III ☐ Non-establishment of report with regard to novelty, inventive step and industrial applicability
 - IV ☐ Lack of unity of invention
 - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
 - VI ☐ Certain documents cited
 - VII ☐ Certain defects in the international application
 - VIII ☐ Certain observations on the international application

Date of submission of the demand 20 February 2001 (20.02.2001)	Date of completion of this report 13 January 2002 (13.01.2002)
Name and mailing address of the IPEA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703)305-3230	Authorized officer James Seidleck Telephone No. (703) 308-0657

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.

PCT/US00/16312

I. Basis of the report

1. With regard to the elements of the international application:*

- ☐ the international application as originally filed.
- ☒ the description:
 pages 1-3, 6-9, 11-30 as originally filed
 pages NONE, filed with the demand
 pages 4, 5 and 10, filed with the letter of 01 October 2001 (01.10.2001)
- ☒ the claims:
 pages NONE, as originally filed
 pages NONE, as amended (together with any statement) under Article 19
 pages NONE, filed with the demand
 pages 31-34, filed with the letter of 01 October 2001 (01.10.2001)

- ☐ the drawings:
 pages NONE, as originally filed
 pages NONE, filed with the demand
 pages NONE, filed with the letter of _____.

- ☐ the sequence listing part of the description:
 pages NONE, as originally filed
 pages NONE, filed with the demand
 pages NONE, filed with the letter of _____.

2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language _____ which is:

- ☐ the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of the translation furnished for the purposes of international preliminary examination (under Rules 55.2 and/or 55.3).

3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in printed form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. ☐ The amendments have resulted in the cancellation of:

- ☐ the description, pages NONE
- ☐ the claims, Nos. NONE
- ☐ the drawings, sheets/fig NONE

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).**

* Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17).

** Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

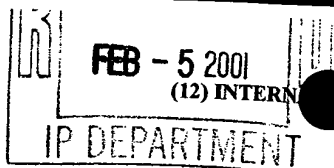
International application No.
PCT/US00/16312**V. Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement****1. STATEMENT**

Novelty (N)	Claims <u>1-23</u>	YES
	Claims <u>NONE</u>	NO
Inventive Step (IS)	Claims <u>1-23</u>	YES
	Claims <u>NONE</u>	NO
Industrial Applicability (IA)	Claims <u>1-23</u>	YES
	Claims <u>NONE</u>	NO

2. CITATIONS AND EXPLANATIONS

Claims 1-23 meet the criteria set out in PCT Article 33(2)-(4), because the prior art does not teach or fairly suggest a resin reinforced silicone composition curable by exposure to radiation or exposure to moisture or a combination, thereof, comprising (a) a polyorganosiloxanes with reactive groups selected from moisture curable groups, photocurable groups, or combinations thereof; (b) a silicone resin selected from the resins in claim 1 or 2; and (c) a cure catalyst selected from the groups consisting of photoinitiators, moisture cure catalyst of combinations thereof. Nor does the prior art teach a method of curing the above compositions to form an elastomer comprising the steps of claims 21-23. The prior art document (D1) fails to teach the silicone resins as defined and used in applicant's invention which, are three-dimensional structures verses the linear silicone fluids taught in D1. Additionally, D1 fails to teach using said silicone fluids to reinforce polyorganosiloxanes. The compositions and methods taught by the instant invention can be useful as a conformal coating.

----- NEW CITATIONS -----



FEB - 5 2001

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APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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C08F 2/46,

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60/144,257 19 July 1999 (19.07.1999) US

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(71) Applicant (*for all designated States except US*): LOCTITE CORPORATION [US/US]; 1001 Trout Brook Crossing, Rocky Hill, CT 06067 (US).

Published:

- With international search report.
- Before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments.

(72) Inventor; and

(75) Inventor/Applicant (*for US only*): CHU, Hsien-Kun [US/US]; 6 Harvest Hill, Wethersfield, CT 06109 (US).

(74) Agent: BAUMAN, Steven, C.; Loctite Corporation, 1001 Trout Brook Crossing, Rocky Hill, CT 06067 (US).

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

WO 01/05846 A1

(54) Title: RESIN-REINFORCED UV, MOISTURE AND UV/MOISTURE DUAL CURABLE SILICONE COMPOSITIONS

(57) Abstract: This invention relates to resin-reinforced silicone compositions curable upon exposure to radiation in the electromagnetic spectrum, which compositions when cured demonstrate improved elastomeric properties, such as tensile strength, modulus and elongation. The inventive resin-reinforced silicone compositions may alternatively be rendered curable by exposure to moisture. In addition, the inventive composition may be rendered curable by exposure to radiation in the electromagnetic spectrum, and exposure to moisture. The inventive silicone compositions are particularly well suited for use in electronic conformal coating and potting applications, as well as in automotive gasketing applications, pressure sensitive adhesive applications and the like.

- 1 -

RESIN-REINFORCED UV, MOISTURE AND UV/MOISTURE DUAL
CURABLE SILICONE COMPOSITIONS

5

BACKGROUND OF THE INVENTION

Field of the Invention

This invention relates to resin-reinforced silicone
10 compositions curable upon exposure to radiation in the
electromagnetic spectrum, which compositions when cured
demonstrate improved elastomeric properties, such as tensile
strength, modulus and elongation. The inventive resin-
reinforced silicone compositions may alternatively be rendered
15 curable by exposure to moisture. In addition, the inventive
composition may be rendered curable by exposure to radiation
in the electromagnetic spectrum, and exposure to moisture.
The inventive silicone compositions are particularly well
suited for use in electronic conformal coating and potting
20 applications, as well as in automotive gasketing applications,
pressure sensitive adhesive applications and the like.

Brief Description of Related Technology

Silicone elastomers have been used for potting and encapsulating electrical devices, such as integrated circuits, because of their excellent thermal stability, low temperature flexibility and high dielectric strength. They typically provide shock, vibrational and thermal stress protection on fragile electronic components. [See U.S. Patent Nos. 3,933,712; 4,072,635; 4,087,585; 4,271,425; 4,374,967; and D. Dickson, Jr., Proc. Electric/Electronic Intel. Conf., 12, 92 (1975). See also U.S. Patent No. 4,374,967.]

Certain silicones containing (meth)acrylate functional groups may be cured by ultraviolet light ("UV light"). See U.S. Patent Nos. 4,201,808 and 4,348,454, and U.K. Patent Application GB 2039287A, which, though not producing elastomeric materials, describe UV light curing compositions of silicones containing (meth)acrylates for paper release coatings.

Room temperature vulcanization ("RTV") (also called moisture curable) silicone compositions are well known, such as those sold under the ULTRA tradename.

Loctite Corporation has for a number of years sold under the "NUVASIL" tradename a variety of UV and UV/moisture curable silicone compositions -- that is, silicone compositions curable upon exposure to UV light or UV light, followed by exposure to moisture. These compositions generally provide a reactive polyorganosiloxane component, and a photoinitiator component. Where moisture cure capabilities are also desirable, a moisture cure catalyst is also included. The viscosity of the composition, as well as the physical properties of the cured elastomer, may be modified by the inclusion of a non-reactive silicone fluid, such as a polydimethyl siloxane, for instance one terminated with alkyl silyl groups (e.g., trimethyl silyl, hydroxyl dimethyl silyl,

alkoxy dimethylsilyl and the like). See Patent No. 4,528,081 (Lien), the disclosure of which is hereby expressly incorporated herein by reference. See also U.S. Patent Nos. 5,179,134 (Chu) and 5,300,608 (Chu), the disclosure of each of which is hereby expressly incorporated herein by reference.

Other UV and UV/moisture curable silicone compositions are also known. [See U.S. patent Nos. 5,489,622 (Hara) and 5,384,340 (Hara).]

Silicone elastomers, however, are inherently very weak materials due to weak intermolecular van der Waals forces between the siloxane chains. In the past, improved strength has been imparted to certain of these compositions by including a fumed silica as a reinforcing filler. See U.S. Patent No. 4,675,346 (Nakos), the disclosure of which is expressly hereby incorporated herein by reference. One drawback to this approach in some instances is that dispensability may become difficult, as oftentimes even a small amount of fumed silica tends to increase viscosity.

In addition, inclusion of such reinforcing filler tends to confer a translucent or "milky" appearance to the silicone compositions. Such an appearance can be disadvantageous in many applications, particularly where cure is to occur through exposure to radiation in the electromagnetic spectrum and/or where clarity of the cured elastomer is a desirable property. Radiation cure may become impeded when the composition has a translucent appearance because the radiation pathway through the composition may become blocked.

Recently, Loctite Corporation made an advance in the field of anaerobically-curable silicone compositions by providing compositions including (a) a silicone fluid formed as the reaction product of a first silane having at least one hydrolyzable functional group, and a second silane having a (meth)acrylic functional group and at least one hydrolyzable

functional group. (b) a (meth)acrylate monomer; and (c) polymerization initiator. See U.S. patent No. 5,605,999 (Chu) and 5,635,546 (Rich), the disclosures of each of which are hereby expressly incorporated herein by reference.

5 Silicone products of the type noted above (sometimes referred to as "MQ" resins) have been used to impart reinforcement properties to cured elastomers of heat-curable silicone compositions without increasing the viscosity of the composition, and while maintaining the clarity of the
10 composition.

 MQ resins generally are copolymers of siloxanes formed from reactive trialkylsilyl ("M") and tetra functional silicate ("Q") structures that can be prepared by either
15 cohydrolyzing silanes containing M and Q units or by silylating inorganic silicates with a trialkylsilyl containing silanes.

 Vinyl- or hydride-containing silanes have been added during preparation to yield MQ resins suitable for use in heat-cure silicone compositions. During heat cure, the vinyl
20 groups on the MQ resin and the silicone fluid polymerize in a crosslinked network with the MQ resin incorporated in the network for reinforcement.

 MQ resins have been used to impart reinforcement properties to room temperature vulcanization ("RTV") silicone
25 compositions, as well. See U.S. Patent No. 5,340,887 and European Patent Document EP 767 216. Here, the use of a resin-polymer system is described where both resin and polymer contain hydroxyl groups and may be crosslinked with the addition of an oxime crosslinker.

30 One drawback to the use of MQ resins to reinforce reaction products of silicone-based compositions is their compatibility with the remaining components of the composition -- that is, phase separation is oftentimes seen to occur. This

results in a compromised shelf life stability for certain one-part silicone-based compositions.

Notwithstanding the state-of-the-technology, it would be desirable to provide resin-reinforced radiation and radiation/moisture curable silicone compositions. Such compositions could advantageously be used in a variety of commercial applications and would benefit from the enhanced strength imparted by the resin reinforcement.

10

SUMMARY OF THE INVENTION

The present invention meets the desires discussed above by providing resin-reinforced silicone compositions capable of curing upon exposure to radiation in the electromagnetic spectrum, such as UV light, and exposure to moisture, such as is found under atmospheric conditions, and a combination thereof:

The compositions include:

(a) a polyorganosiloxane, having photocurable groups, such as (meth)acrylate or glycidoxyl functionality, like methacryloxypropyl, vinyl ether groups and the like and/or moisture curable groups, such as alkoxy or aryloxy groups, like methoxy, acetoxy groups, oximino groups, enoloxyl groups, imido groups, amino groups, and the like.

(b) a silicone resin selected from

(i) those formed from at least one silane within the formula $R^1_m R^2_p Si(X)_{4-(m+p)}$ I, where R^1 is a (meth)acrylate functional group or a hydrolyzable group, and R^2 may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} alkylaryl, or a hydrolyzable group, X is a hydrolyzable group, m is an integer from 1 to 3, and $m+p$ is an integer from 1 to 3;

(i) those formed from at least one silane within the formula $R^3_qSi(X)_{4-q}$ II, where R^3 may be the same as or different from R^2 above and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, and C_{7-18} alkylaryl, and q is an integer from 1 to 3, reacted with at least another silane within the formula of $R^4_rR^5_sSi(X)_{4-(r+s)}$ III, where R^4 and R^5 may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} alkylaryl, r is an integer from 1 to 3, and $r+s$ is an integer from 1 to 3, provided the silicone resin formed contains at least some hydrolyzable group, X ; and combinations thereof; and

(c) a photoinitiator, moisture cure catalyst, and combinations thereof.

In a particularly desirable aspect of the invention, the compositions include:

(a) a polyorganosiloxane, having photocurable groups, such as (meth)acrylate functionality, like methacryloxypropyl, and/or moisture curable groups, such as alkoxy or aryloxy groups, like methoxy;

(b) a silicone resin formed as the reaction product of:

(i) at least one silane within the formula $R^1_mR^2_pSi(X)_{4-(m+p)}$ I, where R^1 is a (meth)acrylate functional group or a hydrolyzable group, and R^2 may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} arylalkyl, C_{7-18} alkylaryl, or a hydrolyzable group, X is a hydrolyzable group, m is an integer from 1 to 3, and $m+p$ is an integer from 1 to 3, and

(ii) at least one other silane within the formula $R_nSi(X)_{4-n}$ IV, where R may be the same or different and may be selected from monovalent ethylenically unsaturated

radicals, hydrogen, C₁₋₁₂ alkyl, C₆₋₁₂ aryl, arylalkyl, C₇₋₁₈ alkylaryl, haloalkyl, and haloaryl, X is a hydrolyzable functionality, and n is an integer of from 0 to 3; and

(c) a photoinitiator, a moisture curing catalyst, and combinations thereof.

The invention further provides a process for preparing reaction products from the compositions of the present invention, the steps of which include applying the composition to a desired substrate surface, particularly one having shadow areas not readily accessible to light, and irradiating the coated substrate surface to conditions which are appropriate to effect cure thereof -- e.g., exposure to radiation in electromagnetic spectrum. Optionally, the composition may then be exposed to moisture to further cure the composition.

Also, the invention of course provides the reaction products so-formed by the above-described process, which reaction products demonstrate improved physical properties, such as tensile strength, modulus and elongation.

The present invention will be more fully appreciated by a reading of the detailed description and the illustrative examples which follow thereafter.

DETAILED DESCRIPTION OF THE INVENTION

As noted above, the compositions include:

(a) a polyorganosiloxane, having photocurable groups, such as (meth)acrylate functionality, like methacryloxypropyl, and/or moisture curable groups, such as alkoxy or aryloxy groups, like methoxy;

(b) a silicone resin selected from

(i) those formed from at least one silane within the formula $R^1_m R^2_p Si(X)_{4-(m+p)}$ I, where R¹ is a (meth)acrylate functional group or a hydrolyzable group, and R²

may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C₁₋₁₂ alkyl, C₆₋₁₂ aryl, C₇₋₁₈ alkylaryl, or a hydrolyzable group, X is a hydrolyzable group, m is an integer from 1 to 3, and m+p is an integer from 1 to 3;

(ii) those formed from at least one silane within the formula $R^3_qSi(X)_{4-q}$ II, where R³ may be the same as or different from R² above and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C₁₋₁₂ alkyl, C₆₋₁₂ aryl, C₇₋₁₈ alkylaryl, and q is an integer from 1 to 3, reacted with at least another silane within the formula of $R^4_rR^5_sSi(X)_{4-(r+s)}$ III, where R⁴ and R⁵ may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C₁₋₁₂ alkyl, C₆₋₁₂ aryl, C₇₋₁₈ alkylaryl, r is an integer from 1 to 3, and r+s is an integer from 1 to 3, provided the silicone resin form contains at least some hydrolyzable group, X; and combinations thereof; and

(c) a photoinitiator, a moisture catalyst, and combinations thereof.

In a particularly desirable aspect of the invention, the compositions include:

(a) a polyorganosiloxane, having photocurable groups, such as (meth)acrylate functionality, like methacryloxypropyl, and/or moisture curable groups, such as alkoxy or aryloxy groups, like methoxy;

(b) a silicone resin formed as the reaction product of:

(i) at least one silane within the formula $R^1_mR^2_pSi(X)_{4-(m+p)}$ I, where R¹ is a (meth)acrylate functional group or a hydrolyzable group, and R² may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C₁₋₁₂ alkyl, C₆₋₁₂ aryl, C₇₋₁₈ arylalkyl, C₇₋₁₈ alkylaryl, or a hydrolyzable group, X is a

hydrolyzable group, m is an integer from 1 to 3, and $m+p$ is an integer from 1 to 3, and

(ii) at least one other silane within the formula $R_nSi(X)_{4-n}$ **IV**, where R may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} arylalkyl, C_{7-18} alkylaryl, haloalkyl, and haloaryl, X is a hydrolyzable functionality, and n is an integer of from 0 to 3; and

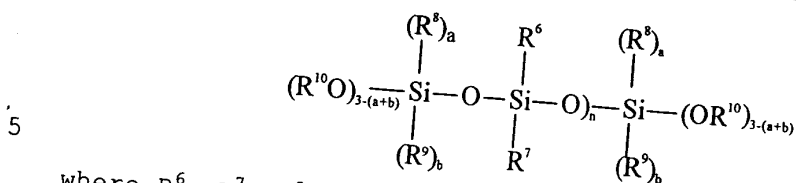
(c) a photoinitiator, a moisture curing catalyst, and combinations thereof.

The inventive compositions may be used as adhesives, coatings, sealants, as well as molding compounds, in applications ranging for instance from preapplied gasketing applications, to virtually any existing RTV silicone application that can accommodate a UV light cure step.

The inventive compositions are capable of being rapidly cured by exposure to UV light, and if desired exposure to atmospheric moisture. Cure typically occurs through exposure to light with a UV intensity of 70,000 mW/cm². The cured compositions are tough elastomeric materials that display typical silicone elastomer properties including wide usable temperature range, and good water resistance.

The polyorganosiloxane should have an average linear molecular size of at least about 50 siloxane units, with (meth)acrylate, (meth)acryloxyalkyldialkoxysilyl and/or (meth)acryloxyalkyldiaryloxysilyl groups on the polyorganosiloxane, such as at the terminal ends. Desirably, the (meth)acryloxyalkyldialkoxysilyl group should be a (meth)acryloxypropyldimethoxysilyl group.

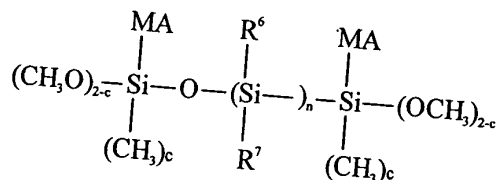
For instance, the polyorganosiloxane may be selected from compounds within formula **V** below:

V

where R^6 , R^7 , R^8 and R^9 may be the same or different and are alkyl, alkenyl, aryl, (meth)acryl, and the like, provided that at least one of R^6 , R^7 , R^8 and R^9 is (meth)acryl, having up to 10 carbon atoms (C_{1-10}), or substituted versions thereof, such as halo- or cyano-substituted; R^{10} is alkyl having up to 10 carbon atoms (C_{1-10}); n is an integer between about 100 and 1,200; a is 1 or 2; b is 0, 1 or 2; and $a+b$ is 1, 2 or 3.

It is particularly desirable to have polyorganosiloxanes, where R^6 and R^7 are methyl, R^8 is (meth)acrylate; R^9 and R^{10} is alkyl, such as methyl, and n is an integer between about 100 and 1,200, inclusive.

Generally, it is convenient to use a linear polyorganosiloxane having (meth)acrylate groups terminating the silicone. Such (meth)acrylate-terminated silicones may be represented by the formula VI below:

VI

where R^6 and R^7 are as defined above, MA is (meth)acrylate and c is 0, 1 or 2.

Generally, the silicone resins are a family of silicone-based materials with a structure represented generally by $(R_3SiO_{1/2})_w(R_2SiO_{2/2})_x(RSiO_{3/2})_y(SiO_{4/2})_z$, where a portion of the total R content includes (meth)acrylate functionality. Desirably, and in the aspect of the invention where at least two silanes form a reaction product, the total

(meth)acrylate containing silicon functionality on the silicone resin may be up to about 15 mole% of the silicone resin, such as in the range of about 1 mole% to about 10 mole%, for instance about 4 mole% to about 8 mole%.

5 Silicone resins may be formed from at least one silane within the formula $R^1_m R^2_p Si(X)_{4-(m+p)}$ I, where R^1 is a (meth)acrylate functional group or a hydrolyzable group, and R^2 may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} arylalkyl, C_{7-18} alkylaryl, or a hydrolyzable group, X is a hydrolyzable functionality, m is an integer of from 1 to 3 and $m+p$ is an integer from 1 to 3, or as reaction products of that at least one silane and at least one second silane within the formula $R_n Si(X)_{4-n}$ II, where R may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} arylalkyl, C_{7-18} alkylaryl, haloalkyl, and haloaryl, X is a hydrolyzable functionality, and n is an integer of from 0 to 3. Certain of these moieties ordinarily may be reaction products of halogenated trialkylsilanes, tetraalkoxysilanes and (meth)acrylate substituted trialkoxysilanes.

The silicone resins may be formed through hydrolysis and condensation reactions.

These silicone resins may also be prepared by (1) acidifying water-soluble sodium silicate, and thereafter capping the resulting sol with a trialkylsilyl group as well as with (meth)acrylate-containing silane or (2) co-hydrolyzing and/or co-condensing silanes containing trialkylsilane groups, vinyl silane groups and tetraalkoxysilane. For a further discussion of silicone resins generally, see R.H. Blaney et al., "Silsesquioxanes", Chem. Rev., 95, 1409-30 (1995).

In the reaction forming such silicone resins, a first silane should be used in an amount within the range of from about 1 to about 10 mole%, such as from about 4 to about

8 mole% of the combination of the first and second silanes. The other silane(s) should be used in an amount with the range of from about 90 to about 99 mole%, such as from about 92 to about 96 mole% of the combination of the first and the other
5 silanes.

In the compositions of the present invention, the hydrolyzable functionality in the silanes may be any functionality which, when attached to a silicon atom through a Si-O, Si-halo, Si-N or Si-S bond, is readily hydrolyzable in
10 the presence of water. Examples of such functionality include, but are not limited to, halogen (meth)acryloxy, alkoxy, aryloxy, isocyanato, amino, acetoxy, oximinioxy, aminoxy, amidato and alkenyloxy.

In the compositions of the present invention, R may
15 be chosen from C₁₋₁₂ alkyl, C₆₋₁₂ aryl, alkenyl, (meth)acryloxyalkyl and vinyl. In such instances, when R is C₁₋₁₂ alkyl or C₆₋₁₂ aryl, examples of the first silane include, but are not limited to, (meth)acryloxypropyl trimethoxysilane, (meth)acryloxypropyl trichlorosilane, (meth)acryloxypropyl
20 dimethylchloro silane, (meth)acryloxymethyl dichlorosilane and (meth)acryloxymethyldimethyl acryloxysilane.

When R¹ on the other silane is chosen from C₁₋₁₂ alkyl, C₆₋₁₂ aryl, the other silane itself may be dimethylchlorosilane, phenyltrichlorosilane,
25 tetrachlorosilane, trimethylchlorosilane, trimethylmethoxysilane, dimethyldimethoxysilane, phenyltrimethoxysilane and tetraethoxysilane.

Of course, appropriate combinations of the first silane may be used as the first silane component; likewise
30 appropriate combinations of the other silane(s) may be used as the other silane component.

The photoinitiator may include any one of those known in the art to initiate the curing of (meth)acrylate functional groups. These photoinitiators include benzoin and

substituted benzophenones, benzophenone, Michler's ketone, dialkoxyacetophenones, such as diethoxyacetophenone, and the like. Generally, the amount of photoinitiator chosen should be within the range of from 0.1 to about 5% by weight.

5 In those compositions in which moisture cure capabilities are also present, a moisture cure catalyst should also be included in an amount effective to cure the composition. For example, from about 0.1 to about 5% by weight, such as about 0.25 to about 2.5% by weight, of the
10 moisture cure catalyst is desirable. Examples of such catalysts include organic titanium, tin, and zirconium complexes and of course combinations thereof. Tetraisopropoxytitanium and tetrabutoxytitanium are particularly desirable. See also U.S. Patent No. 4,111,890, the
15 disclosure of which is expressly incorporated herein by reference.

The compositions may also include a diluent component reactive at elevated temperature conditions.

Reactive diluents include those materials which are
20 particularly (1) unreactive at ambient temperature conditions and (2) reactive at elevated temperature conditions. In addition, such diluents should be capable of not only reacting with other components of the inventive adhesive compositions, but also with reactive moieties on itself. This feature allows
25 the diluent to self-polymerize as well as polymerize with reactive moieties on the other components of the composition. As such, the reactive diluent becomes incorporated into the polymeric matrix which forms at ambient temperature and which further forms at increased temperatures. The incorporation of
30 the reactive diluent provides at least in part for the high temperature performance demonstrated by the cured composition.

More specific examples of such reactive diluents include alkenyl- or alkynyl-terminated silicone fluids, an example of which is vinyl-terminated polydimethylsiloxane.

Other examples of reactive diluents based on silicone fluids include alkenyl- or alkynyl-terminated silicone resins.

And of course appropriate combinations of such reactive diluents may be used.

When used, the reactive diluent should be employed in an amount within the range of about 1 to about 50% by weight, based on the total weight of the composition.

The compositions of the invention may also include other constituents to modify the physical properties of the composition or reaction products thereof, as desired depending on the specific application for which a composition within the scope of this invention is destined for use. For instance, adhesion promoters, such as (meth)acryloxypropyltrimethoxysilane, trialkylisocyanurate, and the like, may be included in an amount of up to about 5% by weight. Conventional silicone fillers, such as fumed and precipitated silica [see e.g. U.S. Patent No. 4,675,346 (Nakos)], iron oxide, barium zirconate and calcium carbonate, may also be included in the inventive compositions. Still other conventional additives may also be included in the inventive compositions, such as non-(meth)acrylate functionalized silicone diluents (including silicone fluids having viscosities of between about 100 and 1,500 cps, which may or may not be terminated with hydrogen, alkyl, alkenyl, alkoxy or hydroxyl functionality, such as trimethylsilyl groups) and plasticizers, each of which may be present in an amount of up to about 30% by weight.

The plasticizers may be chosen from a wide variety of plasticizers depending on the desired properties of the composition and/or reaction product thereof. See e.g., U.S. Patent No. 3,794,610 (Bachmann), the disclosure of which is hereby expressly incorporated herein by reference.

The compositions of the present invention may be prepared using conventional methods that are well known to those persons of skill in the art. For instance, the components of the inventive compositions may be mixed together in any convenient order consistent with the roles and functions the components are to perform in the compositions. Conventional mixing techniques using known apparatus may be employed.

The invention also provides a process for preparing reaction products from the compositions of the present invention, the steps of which include applying the composition to a desired substrate surface and exposing the coated substrate surface to conditions which are appropriate to effect cure thereof -- e.g., exposure to radiation in the electromagnetic spectrum.

Also, the invention of course provides reaction products so-formed by the above-described process, which reaction products demonstrate improved physical properties, such as tensile strength, modulus and elongation.

In view of the above description of the present invention, it is clear that a wide range of practical opportunities is provided.

The following examples are illustrative of the invention, but in no way are intended to limit its scope.

EXAMPLES

1. General Synthesis of (Meth)acrylate Functional Trimethylsilylated Silicates

A. Tetraethoxysilane Method

(Meth)acrylate-functional trimethylsilylated silicate [(meth)acrylate-functional MQ] may be prepared by the co-hydrolysis, co-condensation of trimethylchlorosilane, methacryloxypropyltrimethoxysilane and tetraethoxysilane. As described here, for a MQ resin with an M/Q ratio of about 0.9

and a (meth)acrylate content of about 8 mole% may be prepared. Of course, variations of the M/Q ratio and (meth)acrylate content may be achieved through routine variations in the amount of starting material.

5 In a one-liter, three-neck round bottom flask equipped with a reflux condenser, a mechanical stirrer and an addition funnel was charged with tetraethoxysilane (251.68 g; 1.21 mole), methacryloxypropyltrimethoxysilane (49.60 g; 0.20 mole), trimethylchlorosilane (118.27 g; 1.09 mole) and toluene 10 (150 g). De-ionized water (125 g) was then slowly added to the mixture with stirring through the additional funnel during a period of time of about 15 minutes. The mixture was further heated at reflux for an additional period of time of about three hours, after which the mixture was allowed to cool to 15 room temperature and phase separated in a separatory funnel. The lower layer of the reaction mixture in toluene was collected and returned to the three-neck round bottom flask.

Hexamethyldisilazane (66.0 g) was added to the stirring reaction mixture in toluene through the addition 20 funnel. The mixture was heated at reflux with stirring for a period of time of about three hours, after which the mixture was allowed to cool to room temperature and then filtered. The filtrate was then distilled to remove the toluene-water azeotrope to yield a solution of the MQ resin, whose solids 25 content was determined to be about 50-60%.

B. Sodium Silicate Method

MQ may also be prepared by the reaction of trimethylchlorosilane, methacryloxypropyltrimethoxysilane and 30 sodium silicate. As described here a MQ resin with an M/Q ratio of about 0.6 and a (meth)acrylate content of about 4 mole% may be prepared. Of course, as with the tetraethoxy silane method, variations of the M/Q ratio and (meth)acrylate

content may be achieved through routine variations in the amount of starting material.

A 16.5% hydrochloric acid solution was prepared by mixing 51.03 g of concentrated hydrochloric acid (37%) and 63.41 g of de-ionized water. The acid was then charged into a one-liter three-neck round bottom flask equipped with condenser, mechanical stirrer and addition funnel. The following solutions were then sequentially added to the hydrochloric acid over a period of time of about 5 minutes, with vigorous stirring: (1) aqueous sodium silicate solution prepared by mixing 128.57 g of a 28% silica-containing sodium silicate ($\text{SiO}_2/\text{Na}_2\text{O}=3.22$; 0.60 mole SiO_2) with 183.67 g deionized water; (2) isopropyl alcohol (122.92 g); and (3) a solution of trimethylchlorosilane (39.06 g; 0.36 mole) and methacryloxypropyltrimethoxysilane (9.92 g; 0.04 mole) in toluene (10 g).

After the addition was complete, the mixture was heated to reflux for a period of time of about 3 hours, after which time toluene (150 g) was added. The mixture was allowed to cool to room temperature and phase separated in a separatory funnel. About 190 g of the top layer was collected and placed in a 500 ml, three-neck round bottom flask equipped with a reflux condenser, a mechanical stirrer and an additional funnel.

Hexamethyldisilazane (38 g) was added to the stirring reaction mixture to the solution through the addition funnel. The mixture was heated at reflux for a period of time of about 3 hours and was then allowed to cool to room temperature. The mixture was then filtered, and the solids content determined to be 28%.

2. Synthesis of UV and UV/Moisture Curable Silicones

A. UV Curable Silicones

For instance, to prepare the silicone designated as "A" in Table 1, three hundred (300) grams (96% by weight of the reactants) of the silanol terminated polydimethylsiloxane fluid (having a viscosity of 750 cps and a molecular weight of 12,000) was placed in a one-liter three-neck round bottom flask and mixed with 12.6 g (4% by weight of the reactants) of acryloxymethyldimethylacryloxysilane at room temperature. The mixture was heated under vacuum to remove the acrylic acid by-product formed.

10

B. UV/Moisture Curable Silicones

For instance, to prepare the silicone designated as "C" in Table 1, three hundred (300) grams (95.6% by weight of the reactants) of the silanol terminated polydimethylsiloxane fluid (having a molecular weight of 12,000) was placed in a one-liter three-neck round bottom flask. To the flask were further added 13.64 g (4.4% by weight of the reactants) of methacryloxpropyltrimethoxysilane and 0.25 ml (negligible amount) of 1.6M n-butyl lithium in hexane. The mixture was stirred at room temperature for 30 minutes with nitrogen sparge to obtain a clear fluid. Several pieces of dry ice were then added to the mixture to quench the lithium catalyst.

3. General Preparation of Resin-Reinforced UV and UV/Moisture Curable Silicones

To prepare a resin-reinforced UV curable silicone composition in accordance with the present invention, the solids content of the resin in a toluene solution was first determined as follows. An aluminum dish filled with one gram of the silicone resin/toluene solution was heated at a temperature within the range of about 100 to about 150C. Such heating causes the toluene to evaporate leaving a solid at a constant weight. For a 50% filled composition, the weight of the solution containing 25 g of the resin was then mixed with

25

30

25 g of a UV-curable silicone fluid (for instance, an acryloxymethyl-terminated polydimethylsiloxane, as described in U.S. Patent No. 5,179,134) or a UV/moisture dual cure silicone fluid (for instance, a

- 5 methacryloxypropyldimethoxysilyl-terminated polydimethylsiloxane, as described in U.S. Patent No. 5,663,269). Of course, different levels of filling may also be achieved by varying the amount of resin and polydimethylsiloxane chosen.

- 10 The mixed polymer-resin solution was subjected to rotary vacuum stripping to remove the toluene solvent for a time sufficient to reach a weight of about 50 g. To the stripped mixture was further added diethoxyacetophenone ("DEAP", as a photoinitiator) in an amount of 1.5% by weight.
- 15 In the event that a moisture cure mechanism is also desired, a moisture cure catalyst, such as tetraisopropyltitanium in an amount of 0.5% by weight, should be added.

Examples of UV and UV/moisture silicone compositions, certain of which being filled with silicone resins for comparative purposes, are given below in Table 1.

Table 1

Sample No.	Resin	M/Q Ratio	MA %	UV Silicone	Resin: Siloxane Ratio
1	—	—	—	A	—
2	—	—	—	B	—
3	—	—	—	C	—
4	M4Q6	0.667	0	A	1:1
5	M4Q6	0.667	0	B	1:1
6	M fluid			A	1:1
7	M fluid			B	1:1

* The following conventions are used for resin structures: M for $\text{Me}_3\text{SiO}_{1/2}$; T for $\text{MeSiO}_{3/2}$; T' or T^{Ma} for $\text{CH}_2=\text{C}(\text{Me})\text{COOCH}_2\text{CH}_2\text{CH}_2\text{SiO}_{3/2}$; and Q for $\text{SiO}_{4/2}$.

25

Silicone A is a linear acrylate-terminated polydimethylsiloxane prepared by condensation of a hydroxyl-terminated polydimethylsiloxane having a weight average

molecular weight of 12,000 with acryloxymethyldimethylacryloxysilane. This silicone is used as Sample No. 1. Silicone B is an acrylate-terminated polydimethylsiloxane silicone prepared as above, though from a hydroxyl-terminated polydimethylsiloxane having a weight average molecular weight of 28,000. This silicone is used as Sample No. 2. Silicone C is a methacryloxypropyldimethoxy-terminated polydimethylsiloxane having a weight average molecular weight of 20,000. This silicone is used as Sample No. 3.

Sample Nos. 4 and 5 were prepared by mixing silicone A and B with a non-functional MQ resin. Sample Nos. 6 and 7 were prepared by mixing silicone A and B with a trimethyl-terminated polydimethylsiloxane (having a viscosity of 100 cps).

4. Cure of UV Curable MQ-Reinforced Silicones

The samples as described in Table 1 were cured into 75 mil films by exposure for one minute on each side to UV light emitted from a medium pressure mercury lamp with an intensity of 70 mW/cm² (at 365 nm). Where moisture cure was also desirable, the cured films were allowed to stand at ambient conditions for additional time (such as at least about three to about seven days) before physical properties of cured films were determined.

5. Physical Properties of Unfilled and Inert Additive Filled, UV Silicones

In order to establish comparative data, physical properties of cured silicone compositions with and without an inert additive were determined, including tensile strength, modulus at 50% elongation and elongation at rupture of test pieces, previously cut from cured slabs of the compositions.

Table 2 below shows that the unfilled UV silicones

-- Sample Nos. 2 and 3 -- when cured demonstrated poor physical properties. The tensile strength and elongation of the cured silicone elastomers were low: between 37-53 psi and between 38-71%, respectively. Two inert additives -- a silanol resin prepared by cohydrolyzing trimethylchlorosilane and tetraethoxysilane in a mole ratio of 4:6, and a trimethylsilyl-terminated polydimethylsiloxane with a viscosity of 100 cps -- were also added at a 1:1 ratio to the acrylate-terminated polyorganosiloxanes A and B. These are Sample Nos. 4-5 and 6-7, respectively.

Table 2

Sample No.	Physical Properties		
	Tensile (psi)	Modulus (@ 50%)	Elongation (%)
1	37	—	71
2	53	—	38
3	42	—	63
4	25	10	103
5	60	7	206
6	17	13	68
7	88	10	230

From a comparison of the data presented in Table 2, it may be seen that the silicone elastomers with inert additives demonstrated weak tensile properties, thereby showing very little reinforcement and modulus. The higher elongation of elastomers with additives can be attributed to the plasticizing effect of the additives.

**6. Reinforcement of UV Curable Silicones
with Various Ratios of Different Resins**

Four silicone resins were prepared for evaluation with various levels of M, T and Q, with the "T" unit representing methacryloxyalkyltrifunctional silyl. These resins may be designated as: $M_{42}T^{MA}_3Q_{55}$ (1), $M_{40.8}T^{MA}_{5.8}Q_{53.4}$ (2), $M_{45}T^{MA}_5Q_{50}$ (3) and $M_{44}T^{MA}_8Q_{48}$ (4), where the numerical subscript represents the mole% of M, T and Q starting material used

during resin preparation, as described in Section 1 above. These resins were mixed together with Silicone B to prepare the compositions to be evaluated.

Resins 1 and 2 had a M/Q ratio of 0.764; Resin 3 had a M/Q ratio of 0.9; and Resin 4 had a M/Q ratio of 0.917.

Resin 2 had a (meth)acrylate content of twice that of Resin 1, and Resin 4 had a (meth)acrylate content 1.6 times that of Resin 3. These resins were each mixed with Silicone B in ratios ranging from 1:2 to 2:1, as described in Section 3 above. The M/Q ratios, (meth)acrylate content and resin/siloxane ratios are given in Table 3a below.

Table 3a

Sample No.	Resin	M/Q Ratio	MA %	Resin: Siloxane Ratio
8	1	0.76	3	1:2
9	1	0.76	3	1:1
10	1	0.76	3	2:1
11	2	0.76	5.8	1:2
12	2	0.76	5.8	1:1
13	2	0.76	5.8	2:1
14	3	0.9	5	1:2
15	3	0.9	5	1:1
16	3	0.9	5	2:1
17	4	0.9	8	1:2
18	4	0.9	8	1:1
19	4	0.9	8	2:1

The resin-reinforced compositions were cured by the method described in Section 4 above. The physical properties of the cured silicone elastomers are given below in Table 3b.

Table 3b

Sample No.	Physical Properties			
	Tensile (psi)	Modulus (@ 50%)	Modulus (@ 100%)	Elongation (%)
8	183	23	37	250
9	235	32	55	212
10	428	65	100	232
11	331	85	197	142
12	622	316	—	88
13	602	386	—	80

14	188	38	72	64
15	418	77	202	164
16	493	178	424	114
17	359	143	330	107
18	452	290	-	79
19	466	-	-	28

Within each resin-reinforced silicone composition, higher levels of resin produced a cured silicone elastomer with higher tensile strength and higher modulus values. In addition, lower elongation values were observed to be present in the cured elastomers when higher levels of resin were used.

7. Resin Reinforcement of UV Curable Silicone Compositions with Siloxanes of Different Molecular Weights

Three silicone resins (1, 3 and 4) were mixed with polyorganosiloxanes A, B and C at different ratios to achieve different levels of resin filler in the silicone compositions. These compositions and the relative components are shown below in Table 4a.

Table 4a

Sample No.	Resin	M/Q Ratio	MA %	UV Silicone	Resin: Siloxane Ratio	Cure Mode
20	1	0.764	3	A	1:1	UV
21	1	0.764	3	C	1:1	UV
22	1	0.764	3	C	1:1	dual
23	1	0.764	3	B	1:1	UV
24	3	0.9	5	A	2:1	UV
25	3	0.9	5	A	1:1	UV
26	3	0.9	5	A	1:2	UV
27	3	0.9	5	B	2:1	UV
28	3	0.9	5	B	1:1	UV
29	3	0.9	5	B	1:2	UV
30	4	0.9	8	A	1:1	UV
31	4	0.9	8	C	1:1	UV
32	4	0.9	8	C	1:1	dual
33	4	0.9	8	B	1:1	UV

For Silicone C, UV cure (with photoinitiator added but no moisture curing catalyst present) as well as UV/moisture dual cure conditions (both photoinitiator and moisture curing catalyst present; UV cure followed by at least

2 days additional moisture cure) were used to cure the siloxane into an elastomer.

These resin-filled silicone compositions were prepared and cured, as described in Sections 2 and 3 above.

5 The physical properties of the cured elastomers are shown below in Table 4b.

Table 4b

Sample No.	Tensile (psi)	Modulus (@ 50%)	Modulus (@ 100%)	Elongation %
20	221	55	161	110
21	218	37	79	140
22	432	62	121	180
23	235	32	55	212
24	464	209	-	79
25	282	92	-	93
26	62	51	-	55
27	493	178	424	114
28	418	77	202	164
29	188	38	72	164
30	321	-	-	34
31	656	480	-	60
32	803	677	-	59
33	452	290	-	79

The three silicone resins evaluated in the UV curable silicone compositions showed that elongation of the cured elastomer increases as does the molecular weight of the siloxane. Modulus of the cured elastomers on the other hand appears to decrease with an increase of the molecular weight of the siloxane. Comparison of silicone compositions (Sample Nos. 25-30) prepared from Silicones B and C with Resin 3 [M/Q=0.9; 5% (meth)acrylate] shows that the overall strength of the cured elastomer appears to be a function of the resin:siloxane ratios. Comparing Sample Nos. 21 and 22 with Sample Nos. 31 and 32, it may be seen that UV cure followed by moisture cure imparts improved tensile strengths as well as increased modulus to the cured elastomers.

8. Silicone Reinforcement with Various Resins

Various silicone resins were prepared and mixed with an acrylate-terminated polydimethylsiloxane having a number average molecular weight of about 28,000 to evaluate the degree of reinforcement provided to the cured elastomer. To simplify the evaluation, the compositions were prepared at resin/polymer mixtures of 1:1 ratios. The tensile strengths, moduli at 50%, and elongation of the cured elastomer were determined and given as tensile (psi)/modulus at 50% (psi)/elongation below in Table 5.

Table 5

M/Q Ratio	MA Content (%) tensile (psi)/modulus @ 50% (psi)/elongation (%)						
	3	3.3	4	5	5.8	8	10
0.616	-	-	182 164 64	-	-	-	-
0.703	-	396 220 98	-	-	-	-	-
0.764	235 32 212	-	-	-	622 316 88	-	-
0.9	-	-	-	535 128 168	-	452 290 79	-
1.205	4 1 179	-	-	-	-	-	-
1.25	-	-	-	-	-	-	388 261 86

M/Q ratio gives a relative measure of the molecular weight of the resin. A resin with a low M/Q ratio tends to be of higher molecular weight (and therefore more viscous), and a resin with high M/Q ratios tend to be of low molecular weight (and therefore less viscous). Resins with high M/Q ratios tend to be more compatible with UV silicones and possess lower viscosities.

On the other hand, the number of polymerizable groups on the resin itself is determined by its total methacrylate content as well as its M/Q ratio. Thus, for two

resins with the same methacrylate content, resin having a lower M/Q ratio will have a higher number of methacrylate per resin molecule due to the total methacrylate groups being evenly distributed to fewer molecules. This is due to higher molecular weight associated with a lower M/Q ratio. As a consequence, resins having low M/Q ratios but high methacrylate contents may offer better reinforcement.

In Table 5, it is seen that the resin with high M/Q (1.205) and low methacrylate content (3%) did not confer reinforcement to the cured elastomer. This is due to the low methacrylate content on the resin. Increasing methacrylate (say up to 10%) in a resin with a still higher M/Q ratio (say 1.25) leads to reinforcement as shown in Table 5. However, with such a high methacrylate content, the cured elastomer exhibited a milky white appearance indicating separate resin and siloxane domains.

Although resins with wide ranges of M/Q ratios and (meth)acrylate contents were found to reinforce UV and/or UV/moisture curable silicone compositions, particularly desirable combinations offering excellent reinforcement and dispensability have MQ ratios within the range of about 0.7 to 0.9, and (meth)acrylate contents within the range of about 4-8%.

9. Moisture Curable Resin Reinforced Moisture Curable Silicones

A moisture curable resin reinforced silicone formulation was prepared by mixing 352 g of a MQOH-1 resin (50% solid in xylene with 1% silanol, available from PCR, Gainesville, Florida) with 250 g of a silanol-terminated polydimethylsiloxane with a molecular weight of 12,000. To this mixture was further added 50.74 g of vinyltrimethoxysilane. One ml of a 1.6M n-butyllithium was then added and the mixture was heated to distill away the methanol that was formed from capping of the

vinyltrimethoxysilane to the silanol. After distillation of the methanol, the mixture was further vacuum stripped to remove the xylene solvent to yield a 450 g of a clear liquid.

The liquid was catalyzed by the addition of 0.5% by weight of dibutyltin dilaurate. The formulation was then allowed to cure under ambient conditions for 4 days (Sample No. 34) and 7 (Sample No. 35) days. The physical properties of the cured elastomers are shown below in Table 6.

Table 6

Sample No.	Tensile (psi)	Modulus (@50%)	Modulus (@100%)	Modulus (@200%)	Elongation (%)
34	228	43	56	86	393
35	283	57	74	117	349

A typical unfilled room temperature vulcanizing formulation would exhibit physical properties with less than 100 psi tensile and less than 100% elongation under these conditions, much like the physical properties shown by unfilled Sample Nos. 1-3.

**10. Moisture Curable Resin Reinforced
UV/Moisture Curable Silicones**

A resin with moisture cure capability was prepared by reacting 500 g of MQOH-1 (commercially available from PCR; 50% solids in xylene) with 60 g of vinyltrimethoxysilane using 1 ml of a 1.6M n-butyllithium. The mixture was distilled at a temperature of about 60°C to remove approximately 60 g of methanol. The solids content of the resulting resin was determined by heating 1.00 g thereof in an aluminum dish on a hot plate until constant weight was reached. It was found to be 60%.

To a 250 g of the resin was added 135 g of a methacryloxypropyldimethoxysilyl-capped polydimethylsiloxane with a molecular weight of 12,000. The mixture was further

heated under vacuum to remove the xylene solvent to yield 285 g of a clear liquid. Diethoxyacetophenone (2.85 g) and tetraisopropyltitanate (1.43 g) were further added to the mixture.

5 The catalyzed mixture was exposed to UV radiation to form cured silicone elastomers. The physical properties of the initially UV cured elastomer (Sample No. 36) as well as elastomers cured by exposure to UV radiation, followed by exposure to atmospheric moisture for 3 days (Sample No. 37), 7 days (Sample No. 38) and 10 days (Sample No. 39) were
10 determined and are shown below in Table 7.

Table 7

Sample No.	Tensile (psi)	Modulus (@50%)	Modulus (@100%)	Modulus (@200%)	Elongation (%)
36	76	25	40	70	216
37	317	117	183	--	180
38	441	136	215	425	207
39	468	149	237	--	198

15 These results indicate that the initially UV cured silicone elastomer was weak, and lacked reinforcement, while UV cured silicone elastomers subsequently moisture cured through both the resin and the UV crosslinked polydimethylsiloxane rendered the silicone elastomers
20 reinforced with high tensile strengths and high moduli.

11. UV/Moisture Curable Resin Reinforced UV/Moisture Curable Silicones

25 A UV/moisture curable resin was prepared by reacting 150 g of MQOH-1 with 26.26 g of methacryloxypropyltrimethoxysilane in the presence of 0.3 ml of a 1.6M n-butyllithium. Methanol thus formed from the reaction was distilled away from the mixture. The solids
30 content of the capped resin was found to be 52%.

Fifty grams of this capped resin solution was mixed with 26 g of a methacryloxypropyldimethoxysilyl-terminated polydimethylsiloxane with a molecular weight of 12,000. The resin-polymer mixture in solution was further catalyzed with 0.5 g of diethoxyacetophenone and 0.25 g of tetraisopropyltitanate. A UV cured sample was observed to be fragile and therefore its physical properties were not obtained. However, Sample No. 40 was moisture cured with simultaneous solvent evaporation for 7 days, and Sample No. 41 was UV cured followed by further moisture cure and simultaneous solvent evaporation for 7 days.

The process was repeated with a higher molecular weight UV/moisture curable silicone. Thus, 50 g of the capped resin solution was mixed with 26 g of a methacryloxypropyldimethoxysilyl-terminated polydimethylsiloxane with a molecular weight of 20,000. The resin-polymer mixture in solution was also catalyzed with 0.5 g of diethoxyacetophenone and 0.25 g of tetraisopropyltitanate. A UV cured sample of this mixture was also observed to be fragile and therefore its physical properties were not obtained. Sample No. 42 was UV cured followed by 7 days of further moisture cure with solvent evaporation, and resulted in a strong silicone elastomer. The physical properties of Sample Nos. 40-42 are shown below in Table 8.

Table 8

Sample No.	Mode of Cure	Tensile (psi)	Modulus (@50%)	Modulus (@100%)	Modulus (@200%)	Elongation (%)
40	Moisture	302	60	89	148	392
41	UV/moisture	425	379	--	--	56
42	UV/moisture	500	273	--	--	85

These results indicate that moisture cure only of the system leads to the formation of a reinforced rubber with high elongation whereas UV/moisture dual cure results in the

formation of strong elastomers with tighter crosslinking than moisture cure alone. Compare these results with Sample No. 3, reinforcement due to moisture cure and UV/moisture cure were evident.

5 A UV curable silicone may be formulated with a UV/moisture curable resin to yield an elastomer cured by both UV and moisture, having beneficial properties.

10 Though these examples have been provided for illustrative purposes, the true measure of the spirit and scope of the invention is as set forth in the claims.

What is Claimed is:

1. A resin-reinforced silicone composition curable by exposure to radiation in the electromagnetic spectrum, exposure to moisture and combinations thereof, comprising:

(a) a polyorganosiloxane with reactive groups selected from the group consisting of photocurable groups, moisture curable groups, and combinations thereof;

(b) a silicone resin selected from

(i) those formed from at least one silane within the formula $R^1_m R^2_p Si(X)_{4-(m+p)}$ I, where R^1 is a (meth)acrylate functional group or a hydrolyzable group, and R^2 may be the same or different and may be selected from the group consisting of monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} alkylaryl, and hydrolyzable groups, X is a hydrolyzable group, m is an integer from 1 to 3, and $m+p$ is an integer from 1 to 3;

(ii) those formed from at least one silane within the formula $R^3_q Si(X)_{4-q}$ II, where R^3 may be the same as or different from R^2 above and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} alkylaryl, and q is an integer from 1 to 3, reacted with at least another silane within the formula of $R^4_r R^5_s Si(X)_{4-(r+s)}$ III, where R^4 and R^5 may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} alkylaryl, r is an integer from 1 to 3, and $r+s$ is an integer from 1 to 3, provided the silicone resin formed contains at least some hydrolyzable group, X ; and combinations thereof; and

(c) a cure catalyst selected from the group consisting of a photoinitiator component; a moisture curing catalyst, and combination thereof.

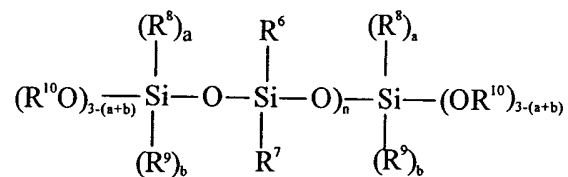
2. The composition of claim 1, wherein the silicone resin is formed as a reaction product of the at least

one silane with the formula $R^1_m R^2_p Si(X)_{4-(m+p)}$, and at least one other silane within the formula, $R_n Si(X)_{4-n}$ **IV**, wherein R may be the same or different and selected from the group consisting of hydrogen, C₁₋₁₂ alkyl, C₆₋₁₂ aryl, C₇₋₁₈ arylalkyl, C₇₋₁₈ alkylaryl, haloalkyl, and haloaryl, X is a hydrolyzable functionality, and n is an integer from 0 to 3.

3. The composition of Claim 1, wherein the photocurable group is selected from the group consisting of acrylate, methacrylate and glycidoxyl groups.

4. The composition of Claim 3, wherein the acrylate group is acryloxypropyl and the methacrylate group is methacryloxypropyl.

5. The composition of Claim 1, wherein the polyorganosiloxane is within the formula:



wherein R⁶, R⁷, R⁸ and R⁹ may be the same or different and are alkyl, alkenyl, aryl, and (meth)acryl, provided that at least one of R⁶, R⁷, R⁸ or R⁹ is (meth)acryl, having up to 10 carbon atoms (C₁₋₁₀), or substituted versions thereof; R¹⁰ is alkyl having up to 10 carbon atoms; n is an integer between about 150 and 1,200; a is 1 or 2; b is 0, 1 or 2; and a+b is 1, 2 or 3.

6. The composition of Claim 1, wherein the moisture curable group is an alkoxy group.

7. The composition of Claim 1, wherein the photoinitiator component is selected from the group consisting of substituted or unsubstituted benzoin, benzophenone,

dialkoxybenzophenones, Michler's ketone, diethoxyacetophenone and combinations thereof.

8. The composition of Claim 6, wherein the photoinitiator component is diethoxyacetophenone.

9. The composition of Claim 1, wherein the moisture curing catalyst is selected from the group consisting of organic titanium derivatives, organic tin derivatives and combinations thereof.

10. The composition of Claim 1, wherein the photocurable group is reactive when exposed to elevated temperature conditions.

11. The composition of Claim 1, further comprising a reactive diluent component.

12. The composition of Claim 11, wherein the reactive diluent is a member selected from the group consisting of vinyl-terminated polydimethylsiloxane, vinylterminated silicone resin, and combinations thereof.

13. The composition of Claim 2, wherein the at least one first silane is present in an amount within the range of from about 1 to about 10 mole% and the at least one other silane is present in an amount within the range of from about 90 to about 99 mole%, of the total of the silanes.

14. The composition of Claim 1, wherein the polyorganosiloxane is present in an amount within the range of from about 15 to about 85 mole% of the total composition.

15. The composition of Claim 1, wherein the photoinitiator component is present in an amount within the range of from about 0.1 to about 5 mole% of the total composition.

16. The composition of Claim 1, wherein the moisture curing catalyst is present in an amount within the range of from about 0.1 to about 5 mole% of the total composition.

17. Irradiated reaction products of the composition of anyone of Claims 1-16.

18. Moisture cure reaction products of the composition of any one of Claims 1-16.

19. The reaction products of Claim 17, further cured by moisture.

20. The reaction products of any one of Claims 17-19, further cured by heat.

21. A method of curing a silicone composition to form an elastomer, the steps of which comprise:

a. applying a silicone composition of any one of Claims 1-16 to a substrate; and

b. irradiating the substrate with UV irradiation sufficient to substantially cure the composition.

22. A method of curing a silicone composition to form an elastomer, the steps of which comprise:

a. applying a silicone composition of any one of Claims 1-16 to a substrate; and

b. exposing the composition to moisture sufficient to substantially cure the composition.

23. A method of curing a UV/moisture dual curing silicone composition to form an elastomer, the steps of which comprise:

a. applying a silicone composition of any one of Claims 1-16 to a substrate having shadow areas not readily accessible to ultraviolet or visible radiation in an amount sufficient to coat the ultraviolet and visible radiation-accessible and the shadow areas;

b. irradiating the substrate with UV irradiation sufficient to substantially cure the composition in the ultraviolet and visible radiation-accessible area; and

c. exposing the composition on the substrate to moisture for a time sufficient to cure the composition in the shadow areas.

meth)acrylic functional group and at least one hydrolyzable functional group. (b) a (meth)acrylate monomer; and (c) polymerization initiator. See U.S. Patent No. 5,605,999 (Chu) and 5,635,546 (Rich), the disclosures of each of which are
5 hereby expressly incorporated herein by reference.

Silicone products of the type noted above (sometimes referred to as "MQ" resins) have been used to impart reinforcement properties to cured elastomers of heat-curable silicone compositions without increasing the viscosity of the
10 composition, and while maintaining the clarity of the composition.

MQ resins generally are copolymers of siloxanes formed from reactive trialkylsilyl ("M") and tetra functional silicate ("Q") structures that can be prepared by either
15 cohydrolyzing silanes containing M and Q units or by silylating inorganic silicates with a trialkylsilyl containing silanes.

Vinyl- or hydride-containing silanes have been added during preparation to yield MQ resins suitable for use in
20 heat-cure silicone compositions. During heat cure, the vinyl groups on the MQ resin and the silicone fluid polymerize in a crosslinked network with the MQ resin incorporated in the network for reinforcement.

MQ resins have been used to impart reinforcement
25 properties to room temperature vulcanization ("RTV") silicone compositions, as well. See U.S. Patent No. 5,340,887 and European Patent Document EP 767 216. Here, the use of a resin-polymer system is described where both resin and polymer contain hydroxyl groups and may be crosslinked with the
30 addition of an oxime crosslinker.

One drawback to the use of MQ resins to reinforce reaction products of silicone-based compositions is their compatibility with the remaining components of the composition -- that is, phase separation is oftentimes seen to occur. This

results in a compromised shelf life stability for certain one-part silicone-based compositions.

Notwithstanding the state-of-the-technology, it would be desirable to provide resin-reinforced radiation and radiation/moisture curable silicone compositions. Such compositions could advantageously be used in a variety of commercial applications and would benefit from the enhanced strength imparted by the resin reinforcement.

10

SUMMARY OF THE INVENTION

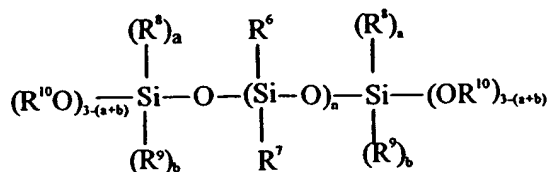
The present invention meets the desires discussed above by providing resin-reinforced silicone compositions capable of curing upon exposure to radiation in the electromagnetic spectrum, such as UV light, and exposure to moisture, such as is found under atmospheric conditions, and a combination thereof:

The compositions include:

(a) a polyorganosiloxane, having photocurable groups, such as (meth)acrylate or glycidoxyl functionality, like methacryloxypropyl, vinyl ether groups and the like and/or moisture curable groups, such as alkoxy or aryloxy groups, like methoxy, acetoxy groups, oximino groups, enoloxo groups, imido groups, amino groups, and the like;

(b) a silicone resin selected from

(i) those formed from at least one silane within the formula $R^1_m R^2_p Si(X)_{4-(m+p)}$ I, where R^1 is a (meth)acrylate functional group or a hydrolyzable group, and R^2 may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} alkylaryl, or a hydrolyzable group, X is a hydrolyzable group, m is an integer from 1 to 3, and $m+p$ is an integer from 1 to 3;

V

5

where R^6 , R^7 , R^8 and R^9 may be the same or different and are alkyl, alkenyl, aryl, (meth)acryl, and the like, provided that at least one of R^6 , R^7 , R^8 and R^9 is (meth)acryl, having up to 10 carbon atoms (C_{1-10}), or substituted versions thereof, such as halo- or cyano-substituted; R^{10} is alkyl having up to 10 carbon atoms (C_{1-10}); n is an integer between about 100 and 1,200; a is 1 or 2; b is 0, 1 or 2; and $a+b$ is 1, 2 or 3.

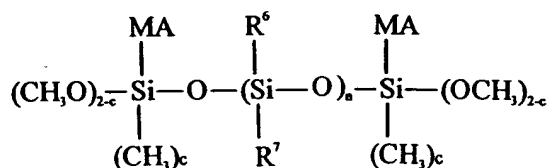
10

It is particularly desirable to have polyorganosiloxanes, where R^6 and R^7 are methyl, R^8 is (meth)acrylate; R^9 and R^{10} is alkyl, such as methyl, and n is an integer between about 100 and 1,200, inclusive.

15

Generally, it is convenient to use a linear polyorganosiloxane having (meth)acrylate groups terminating the silicone. Such (meth)acrylate-terminated silicones may be represented by the formula VI below:

20

VI

where R^6 and R^7 are as defined above, MA is (meth)acrylate and c is 0, 1 or 2.

Generally, the silicone resins are a family of silicone-based materials with a structure represented generally by $(R_3SiO_{1/2})_w(R_2SiO_{2/2})_x(RSiO_{3/2})_y(SiO_{4/2})_z$, where a portion of the total R content includes (meth)acrylate functionality. Desirably, and in the aspect of the invention where at least two silanes form a reaction product, the total

30

What is Claimed is:

1. A resin-reinforced silicone composition curable by exposure to radiation in the electromagnetic spectrum, exposure to moisture and combinations thereof, comprising:

(a) a polyorganosiloxane with reactive groups selected from the group consisting of photocurable groups, moisture curable groups, and combinations thereof;

(b) a silicone resin selected from

(i) those formed from at least one silane within the formula $R^1_m R^2_p Si(X)_{4-(m+p)}$ I, where R^1 is a (meth)acrylate functional group or a hydrolyzable group, and R^2 may be the same or different and may be selected from the group consisting of monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} alkylaryl, and hydrolyzable groups, X is a hydrolyzable group, m is an integer from 1 to 3, and $m+p$ is an integer from 1 to 3;

(ii) those formed from at least one silane within the formula $R^3_q Si(X)_{4-q}$ II, where R^3 may be the same as or different from R^2 above and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} alkylaryl, and q is an integer from 1 to 3, reacted with at least another silane within the formula of $R^4_r R^5_s Si(X)_{4-(r+s)}$ III, where R^4 and R^5 may be the same or different and may be selected from monovalent ethylenically unsaturated radicals, hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} alkylaryl, r is an integer from 1 to 3, and $r+s$ is an integer from 1 to 3, provided the silicone resin formed contains at least some hydrolyzable group, X ; and combinations thereof; and

(c) a cure catalyst selected from the group consisting of a photoinitiator component; a moisture curing catalyst, and combination thereof.

2. A resin-reinforced silicone composition curable by exposure to radiation in the electromagnetic spectrum, exposure to moisture and combinations thereof, comprising:

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(a) a polyorganosiloxane with reactive groups selected from the group consisting of photocurable groups, moisture curable groups, and combinations thereof;

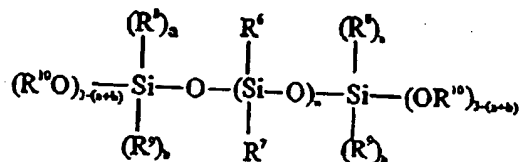
(b) a silicone resin formed as a reaction product of at least one silane within the formula $R^1R^2Si(X)_{4-n}$ I, and at least one other silane within the formula, $R_nSi(X)_{4-n}$ IV, wherein R may be the same or different and selected from the group consisting of hydrogen, C_{1-12} alkyl, C_{6-12} aryl, C_{7-18} arylalkyl, C_{7-18} alkylaryl, haloalkyl, and haloaryl, X is a hydrolyzable functionality, and n is an integer from 0 to 3; and

(c) a cure catalyst selected from the group consisting of a photoinitiator component; a moisture curing catalyst, and combination thereof.

3. The composition of Claim 1 or 2, wherein the photocurable group is selected from the group consisting of acrylate, methacrylate and glycidoxyl groups.

4. The composition of Claim 1 or 2, wherein the acrylate group is acryloxypropyl and the methacrylate group is methacryloxypropyl.

5. The composition of Claim 1 or 2, wherein the polyorganosiloxane is within the formula:



wherein R^6 , R^7 , R^8 and R^9 may be the same or different and are alkyl, alkenyl, aryl, and (meth)acryl, provided that at least one of R^6 , R^7 , R^8 or R^9 is (meth)acryl, having up to 10 carbon atoms (C_{1-10}), or substituted versions thereof; R^{10} is alkyl having up to 10 carbon atoms; n is an integer between about 150 and 1,200; a is 1 or 2; b is 0, 1 or 2; and a+b is 1, 2 or 3.

6. The composition of Claim 1 or 2, wherein the moisture curable group is an alkoxy group.

7. The composition of Claim 1 or 2, wherein the photoinitiator component is selected from the group consisting of substituted or unsubstituted benzoin, benzophenone, dialkoxybenzophenones, Michler's ketone, diethoxyacetophenone and combinations thereof.

8. The composition of Claim 6, wherein the photoinitiator component is diethoxyacetophenone.

9. The composition of Claim 1 or 2, wherein the moisture curing catalyst is selected from the group consisting of organic titanium derivatives, organic tin derivatives and combinations thereof.

10. The composition of Claim 1 or 2, wherein the photocurable group is reactive when exposed to elevated temperature conditions.

11. The composition of Claim 1 or 2, further comprising a reactive diluent component.

12. The composition of Claim 11, wherein the reactive diluent is a member selected from the group consisting of vinyl-terminated polydimethylsiloxane, vinyl-terminated silicone resin, and combinations thereof.

13. The composition of Claim 2, wherein the at least one first silane is present in an amount within the range of from about 1 to about 10 mole% and the at least one other silane is present in an amount within the range of from about 90 to about 99 mole%, of the total of the silanes.

14. The composition of Claim 1 or 2, wherein the polyorganosiloxane is present in an amount within the range of from about 15 to about 85 mole% of the total composition.
15. The composition of Claim 1 or 2, wherein the photoinitiator component is present in an amount within the range of from about 0.1 to about 5 mole% of the total composition.
16. The composition of Claim 1 or 2, wherein the moisture curing catalyst is present in an amount within the range of from about 0.1 to about 5 mole% of the total composition.
17. Irradiated reaction products of the composition of anyone of Claims 1-16.
18. Moisture cure reaction products of the composition of any one of Claims 1-16.
19. The reaction products of Claim 17, further cured by moisture.
20. The reaction products of any one of Claims 17-19, further cured by heat.
21. A method of curing a silicone composition to form an elastomer, the steps of which comprise:
 - a. applying a silicone composition of any one of Claims 1-16 to a substrate; and
 - b. irradiating the substrate with UV irradiation sufficient to substantially cure the composition.
22. A method of curing a silicone composition to form an elastomer, the steps of which comprise:
 - a. applying a silicone composition of any one of Claims 1-16 to a substrate; and
 - b. exposing the composition to moisture sufficient to substantially cure the composition.
23. A method of curing a UV/moisture dual curing silicone composition to form an elastomer, the steps of which comprise:

INTERNATIONAL SEARCH REPORT

 International application No.
PCT/US00/16312

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : C08F 2/46, 2/48, 2/50, 8/00; C08L 83/00, 47/00

US CL : 522/148, 172, 99; 525/100, 105, 106

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. :

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

PLUS, East, west

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5,486,565 A (GENTLE et al) 23 January 1996, see entire document.	1-23
A	US 5,212,211 A (WELCH, II et al) 18 May 1993, see entire document.	1-23
A	US 5,162,460 A (POPA et al) 10 November 1992, see entire document.	1-23
A	US 4,539,367 A (BEERS) 03 September 1985, see entire document.	1-23
A	US 4,528,353 A (LUCAS et al) 09 Jylu 1985, see entire document.	1-23
A	US 4,513,115 A (BEERS) 23 April 1985, see entire document.	1-23

☒ Further documents are listed in the continuation of Box C.
 ☐ See patent family annex.

* Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be of particular relevance	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E" earlier document published on or after the international filing date	"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&"	document member of the same patent family
"O" document referring to an oral disclosure, use, exhibition or other means		
"P" document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search

17 OCTOBER 2000

Date of mailing of the international search report

14 NOV 2000

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US00/16312

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5,905,123 A (CIFUENTES et al) 18 May 1999, see entire document.	1-23
A,P	US 5,952,397 A (FUJIKI et al) 14 September 1999, see entire document.	1-23
Y	US 5,696,209 A (KING et al) 09 December 1997, see entire document	1-23
A	US 5,561,203 A (STRONG et al) 01 October 1996, see entire document.	1-23
Y	US 5,516,812 A (CHU et al) 14 May 1996, see entire document.	1-23
A	US 5,508,360 A (CIFUENTES et al) 16 April 1996, see entire document.	1-23
Y	US 5,498,642 A (CHU et al) 12 March 1996, see entire document.	1-23

The demand must be filed directly with the competent International Examining Authority or, if two or more Authorities are competent, with the one chosen by the applicant. The name or two-letter code of that Authority may be indicated by the applicant on the line below:
IPEA/ US

PCT

CHAPTER II

DEMAND

Under Article 31 of the Patent Cooperation Treaty:
The undersigned requests that the international application specified below be the subject of
International preliminary examination according to the Patent Cooperation Treaty and
hereby elects all eligible States (except where otherwise indicated).

For International Preliminary Examining Authority use only

Identification of IPEA		Date of receipt of DEMAND
Box No. I IDENTIFICATION OF THE INTERNATIONAL APPLICATION		Applicant's or agent's file reference LC-366 PCT
International application No. PCT/US00/16312	International filing date (day/month/year) 07 JULY 2000 (07.07.00)	(Earliest) Priority date (day/month/year) 19 JULY 1999 (19.07.99)
Title of invention RESIN-REINFORCED UV, MOISTURE AND UV/MOISTURE DUAL CURABLE SILICONE COMPOSITIONS		
Box No. II APPLICANT(S)		
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.) LOCTITE CORPORATION 1001 Trout Brook Crossing Rocky Hill, Connecticut 06067 US		Telephone No.: 860.571.5000 Facsimile No.: 860.571.5465 Teleprinter No.:
State (that is, country) of nationality: US	State (that is, country) of residence: US	
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.) CHU, Hsien-Kun 6 Harvest Hill Wethersfield, CT 06019 US		
State (that is, country) of nationality: US	State (that is, country) of residence: US	
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)		
State (that is, country) of nationality:	State (that is, country) of residence:	
<input type="checkbox"/> Further applicants are indicated on a continuation sheet.		

Form PCT/IPEA/401 (first sheet) (July 1998; reprint July 1999)

See Notes to the demand form

Box No. III AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE

The following person is

☒ Agent☐ common representative

and

☒

has been appointed earlier and represents the applicant(s) also for international preliminary examination.

☐

is hereby appointed and any earlier appointment of (an) agent(s)/common representative is hereby revoked.

☐

is hereby appointed, specifically for the procedure before the International Preliminary Examining Authority, in addition to the agent(s)/common representative appointed earlier.

Name and address: *(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)*

BAUMAN, Steven C.
 LOCTITE CORPORATION
 1001 Trout Brook Crossing
 Rocky Hill, Connecticut 06067
 US

Telephone No.:

860.571-5001

Facsimile No.:

860.571.5028

Teleprinter No.:

☐

Address for correspondence: Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.

Box No. IV BASIS FOR INTERNATIONAL PRELIMINARY EXAMINATION**Statement concerning amendments:***

1. The applicant wishes the international preliminary examination to start on the basis of:

☒

the international application as originally filed

the description

☒

as originally filed

☐

as amended under Article 34

the claims

☒

as originally filed

☐

as amended under Article 19 (together with any accompanying statement)

☐

as amended under Article 34

the drawings

☒

as originally filed

☐

as amended under Article 34

2. ☐ The applicant wishes any amendment to the claims under Article 19 to be considered reversed.
3. ☐ The applicant wishes the start of the international preliminary examination to be postponed until the expiration of 20 months from the priority date unless the International Preliminary Examining Authority receives a copy of any amendments made under Article 19 or a notice from the applicant that he does not wish to make such amendments (Rule 69.1(d)). *(This check-box may be marked only where the time limit under Article 19 has not yet expired.)*
- * Where no check-box is marked, international preliminary examination will start on the basis of the international application as originally filed or, where a copy of amendments to the claims under Article 19 and/or amendments of the international application under Article 34 are received by the International Preliminary Examining Authority before it has begun to draw up a written opinion or the international preliminary examination report, as so amended.

Language for the purposes of international preliminary examination:

☒

which is the language in which the international application was filed.

☐

which is the language of a translation furnished for the purposes of international search.

☐

which is the language of publication of the international application.

☐

which is the language of the translation (to be) furnished for the purposes of international preliminary examination.

Box No. V ELECTION OF STATES

The applicant hereby elects all eligible States *(that is, all States which have been designated and which are bound by Chapter II of the PCT)* excluding the following States which the applicant wishes not to elect:

Box No. VI CHECK LIST

The demand is accompanied by the following elements, in the language referred to in Box No. IV, for the purposes of international preliminary examination:

- | | | | |
|--|---|-------|--------|
| 1. translation of international application | : | _____ | sheets |
| 2. amendments under Article 34 | : | _____ | sheets |
| 3. copy (or, where required, translation) of amendments under Article 19 | : | _____ | sheets |
| 4. copy (or, where required, translation) of statement under Article 19 | : | _____ | sheets |
| 5. letter | : | _____ | sheets |
| 6. other (<i>specify</i>) | : | _____ | sheets |

For International Preliminary Examining Authority use only

received not received


<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>

The demand is also accompanied by the item(s) marked below:

- | | |
|--|---|
| 1. <input checked="" type="checkbox"/> fee calculation sheet | 4. <input type="checkbox"/> statement explaining lack of signature |
| 2. <input type="checkbox"/> separate signed power of attorney | 5. <input type="checkbox"/> nucleotide and or amino acid sequence listing in computer readable form |
| 3. <input type="checkbox"/> copy of general power of attorney; reference number, if any: | 6. <input checked="" type="checkbox"/> other (<i>specify</i>): RETURN RECEIPT POSTCARD |

Box No. VII SIGNATURE OF APPLICANT, AGENT OR COMMON REPRESENTATIVE

Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the demand).


 BAUMAN Steven C., Agent
 (Registration No. 33,832)

For International Preliminary Examining Authority use only

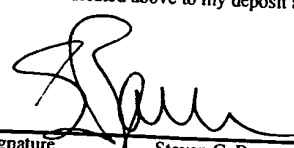
1. Date of actual receipt of DEMAND: _____
2. Adjusted date of receipt of demand due to CORRECTIONS under Rule 60.1(b): _____
3. ☐ The date of receipt of the demand is AFTER the expiration of 19 months from the priority date and item 4 or 5, below, does not apply. ☐ The applicant has been informed accordingly.
4. ☐ The date of receipt of the demand is WITHIN the period of 19 months from the priority date as extended by virtue of Rule 80.5.
5. ☐ Although the date of receipt of the demand is after the expiration of 19 months from the priority date, the delay in arrival is EXCUSED pursuant to Rule 82.

For International Bureau use only

Demand received from IPEA on: _____

FEE CALCULATION SHEET

Annex to the Demand for international preliminary examination

<table border="1" style="width: 100%; border-collapse: collapse;"><tr><td style="width: 50%;">International application No.</td><td style="width: 50%;">PCT/US00/16312</td></tr><tr><td>Applicant's or agent's file reference</td><td>LC-366 PCT</td></tr><tr><td colspan="2">Applicant LOCTITE CORPORATION CHU, Hsien-Kun</td></tr></table>	International application No.	PCT/US00/16312	Applicant's or agent's file reference	LC-366 PCT	Applicant LOCTITE CORPORATION CHU, Hsien-Kun		<table border="1" style="width: 100%; border-collapse: collapse;"><tr><td colspan="2">For International Preliminary Examining Authority use only</td></tr><tr><td colspan="2">Date stamp of the IPEA</td></tr></table>	For International Preliminary Examining Authority use only		Date stamp of the IPEA	
International application No.	PCT/US00/16312										
Applicant's or agent's file reference	LC-366 PCT										
Applicant LOCTITE CORPORATION CHU, Hsien-Kun											
For International Preliminary Examining Authority use only											
Date stamp of the IPEA											
<p>Calculation of prescribed fees</p> <p>1. Preliminary examination fee 490.00 P</p> <p>2. Handling fee (<i>Applicants from certain States are entitled to a reduction of 75% of the handling fee. Where the applicant is (or all applicants are) so entitled, the amount to be entered at H is 25% of the handling fee.</i>) 137.00 H</p> <p>3. Total of prescribed fees Add the amounts entered at P and H and enter total in the TOTAL box 627.00</p> <div style="border: 1px solid black; width: 100px; margin: 0 auto; text-align: center; font-weight: bold;">TOTAL</div>											
<p>Mode of Payment</p> <table style="width: 100%;"><tr><td><input checked="" type="checkbox"/> authorization to charge deposit account with the IPEA (see below)</td><td><input type="checkbox"/> Cash</td></tr><tr><td><input type="checkbox"/> Cheque</td><td><input type="checkbox"/> Revenue stamps</td></tr><tr><td><input type="checkbox"/> postal money order</td><td><input type="checkbox"/> Coupons</td></tr><tr><td><input type="checkbox"/> bank draft</td><td><input type="checkbox"/> Other (specify):</td></tr></table>		<input checked="" type="checkbox"/> authorization to charge deposit account with the IPEA (see below)	<input type="checkbox"/> Cash	<input type="checkbox"/> Cheque	<input type="checkbox"/> Revenue stamps	<input type="checkbox"/> postal money order	<input type="checkbox"/> Coupons	<input type="checkbox"/> bank draft	<input type="checkbox"/> Other (specify):		
<input checked="" type="checkbox"/> authorization to charge deposit account with the IPEA (see below)	<input type="checkbox"/> Cash										
<input type="checkbox"/> Cheque	<input type="checkbox"/> Revenue stamps										
<input type="checkbox"/> postal money order	<input type="checkbox"/> Coupons										
<input type="checkbox"/> bank draft	<input type="checkbox"/> Other (specify):										
<p>Deposit Account Authorization (<i>this mode of payment may not be available at all IPEAs</i>)</p> <p>The IPEA/ <u>US</u> <input checked="" type="checkbox"/> is hereby authorized to charge the total fees indicated above to my deposit account.</p> <p><input checked="" type="checkbox"/> (<i>this check-box may be marked only if the conditions for deposit accounts of the IPEA so permit</i>) is hereby authorized to charge any deficiency or credit any overpayment in the total fees indicated above to my deposit account.</p> <div style="display: flex; justify-content: space-between; margin-top: 20px;"><div style="text-align: center;"><u>12-2135</u> Deposit Account Number</div><div style="text-align: center;"><u>20 February 2001</u> Date (day/month/year)</div><div style="text-align: center;"> Signature Steven C. Bauman</div></div>											

Form PCT/IPEA/401 (Annex) (January 1996; reprint January 1998)

See Notes to the fee calculation sheet

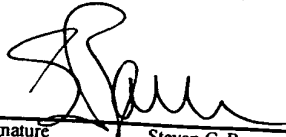
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CHAPTER II

FEE CALCULATION SHEET

Annex to the Demand for international preliminary examination

International application No.	PCT/US00/16312	For International Preliminary Examining Authority use only
Applicant's or agent's file reference	LC-366 PCT	
Applicant LOCTITE CORPORATION CHU, Hsien-Kun		Date stamp of the IPEA
Calculation of prescribed fees		
1. Preliminary examination fee	490.00	P
2. Handling fee (<i>Applicants from certain States are entitled to a reduction of 75% of the handling fee. Where the applicant is (or all applicants are) so entitled, the amount to be entered at H is 25% of the handling fee.</i>)	137.00	H
3. Total of prescribed fees Add the amounts entered at P and H and enter total in the TOTAL box	627.00	
		TOTAL
Mode of Payment		
<input checked="" type="checkbox"/> authorization to charge deposit account with the IPEA (see below)	<input type="checkbox"/> Cash	
<input type="checkbox"/> Cheque	<input type="checkbox"/> Revenue stamps	
<input type="checkbox"/> postal money order	<input type="checkbox"/> Coupons	
<input type="checkbox"/> bank draft	<input type="checkbox"/> Other (specify):	
Deposit Account Authorization (<i>this mode of payment may not be available at all IPEAs</i>)		
The IPEA/ <u>US</u> <input checked="" type="checkbox"/> is hereby authorized to charge the total fees indicated above to my deposit account.		
<input checked="" type="checkbox"/> (<i>this check-box may be marked only if the conditions for deposit accounts of the IPEA so permit</i>) is hereby authorized to charge any deficiency or credit any overpayment in the total fees indicated above to my deposit account.		
<u>12-2135</u> Deposit Account Number	<u>20 February 2001</u> Date (day/month/year)	 Signature Steven C. Bauman

Form PCT/IPEA/401 (Annex) (January 1996; reprint January 1998)

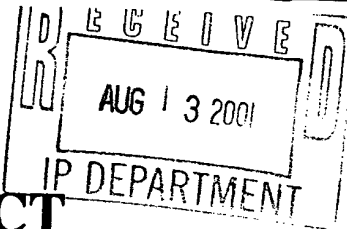
See Notes to the fee calculation sheet

\\LCHQMAIN\APPS\US\src\Bauman\SI\WORD\PATENTS\PCT DEMAND FEE CALC\LC-366.doc

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page 1 of 1

PATENT COOPERATION TREATY



From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:
STEVEN C. BAUMAN
LOCTITE CORPORATION
1001 TROUT BROOK CROSSING
ROCKY HILL, CT 06067

SEARCHED	INDEXED
SERIALIZED	FILED
AUG 15 2001	

PCT

WRITTEN OPINION

(PCT Rule 66)

Applicant's or agent's file reference		Date of Mailing (day/month/year)
LC-366-PCT		10 AUG 2001
International application No.		REPLY DUE
PCT/US00/16312	International filing date (day/month/year)	within 2 months/days from the above date of mailing
	07 July 2000 (07.07.2000)	10/10/01
International Patent Classification (IPC) or both national classification and IPC		Priority date (day/month/year)
IPC(7): C08F 2/48; C08L 67/00; 75/06; C08G 63/00 and US Cl.: 522/60; 24, 66, 71, 81, 104, 105.; 528/280, 281, 282; 525/410, 411, 437, 445		19 July 1999 (19.07.1999)
Applicant		
LOCTITE CORPORATION		

1. This written opinion is the first (first, etc.) drawn by this International Preliminary Examining Authority.
2. This opinion contains indications relating to the following items:
 - I ☒ Basis of the opinion
 - II ☐ Priority
 - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
 - IV ☐ Lack of unity of invention
 - V ☒ Reasoned statement under Rule 66.2 (a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
 - VI ☐ Certain documents cited
 - VII ☐ Certain defects in the international application
 - VIII ☐ Certain observations on the international application

3. The applicant is hereby **invited to reply** to this opinion.

When? See the time limit indicated above. ~~The applicant may, before the expiration of that time limit, request this Authority to grant an extension. See rule 66.2(d).~~

How? By submitting a written reply, accompanied, where appropriate, by amendments, according to Rule 66.3. For the form and the language of the amendments, see Rules 66.8 and 66.9.

Also For an additional opportunity to submit amendments, see Rule 66.4.
For the examiner's obligation to consider amendments and/or arguments, see Rule 66.4 *bis*.
For an informal communication with the examiner, see Rule 66.6

If no reply is filed, the international preliminary examination report will be established on the basis of this opinion.

4. The final date by which the international preliminary examination report must be established according to Rule 69.2 is: 19 October 2001 (19.10.2001)

Name and mailing address of the IPEA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703)305-3230	Authorized officer James Seidleck Telephone No. (703) 308-0657 DEBORAH THOMAS PARALEGAL SPECIALIST
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